Concept for Testing Noble Element Atomic Energy Levels and Photoelectric Cross Sections Using Cyclotron Radiation Emission Spectroscopy

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Tests of theoretical calculations of the energy levels and photoelectric cross-sections for the stable noble elements can be determined with eV-scale accuracy using cyclotron radiation emission spectroscopy (CRES). Proven feasible in 2015, CRES has been previously used to perform high-precision measurements of electron energies from internal radioactive decays of $^{83}\text{m}^\text{Kr}$. We present a concept for a new kind of CRES system which can act as general purpose X-ray detector. The conceptualization includes experimental considerations based on the noble target material, density, electron trapping depth, and noise levels. We exercise these considerations using the example of an argon target irradiated by X-rays from the decay of $^{55}\text{Fe}$, and present an analysis algorithm for spectral deconvolution. We end with a discussion of how the approach could be expanded beyond the noble elements.

I. INTRODUCTION

Modern physics tools and databases involving the X-ray absorption edges for the elements\cite{1-4} are largely based on a small number of publications that are half a century old\cite{5,6}. During the intervening time, these databases have been cross-compared with both theoretical\cite{7} and experimental\cite{8} approaches, though a full review of validations of the databases is beyond the scope of this work.

These cross-checks, however, can have systematic limitations based on disagreements in the underlying theory. For example, the NIST XCOM databases have discrepancies in the interaction cross-sections of up to 15\% far from the absorption edges, and over 50\% near them. On the experimental side, the comparisons are limited by the instrumental resolution of the X-ray detector. The highest-resolution magnetic microcalorimeters have 0.1\% resolution\cite{9}, and X-ray diffraction systems can have resolution on the order of 1 part in $10^7$\cite{10}. These instruments, however, obtain these levels of performance over a very narrow energy range, from order 1 keV for microcalorimeters to order 10 meV for the X-ray diffraction system, and outside this range the system must be re-tuned for a different energy. For more general-purpose, broad-band semiconductor detectors, representative energy resolution is on the order of 1\%\cite{11}.

A relatively new method for measuring electron energies is via cyclotron radiation emission spectroscopy, or CRES. Originally proposed in 2009\cite{12} and proven feasible in 2015\cite{13}, its original purpose was to perform a direct measurement of the mass of the electron anti-neutrino via a high-resolution analysis of the endpoint of the atomic tritium beta spectrum. The prototype detector obtained an instrumental resolution of 2.8 eV\cite{14}, with an optimized system perhaps capable of sub-eV resolution\cite{15}. Unlike microcalorimeters and X-ray diffraction energy measurements, this resolution is anticipated to be independent of energy, allowing for 0.1\% at 1 keV to 0.001\% for transuranic K-shell transitions.

If the CRES technique is applied to an appropriate target, we might perform measurements of both X-ray absorption edges and photoelectric cross-sections in a single system. The effective energy range of the simplest system is on the order of 10 keV, although separating and amplifying the signal line can increase this range to over 100 keV via use of multiple downmixers (see Section IV). Because a single system would be used to probe multiple targets and sources, both absolute and relative measurements could be obtained, with the systematic uncertainties of the latter category greatly reduced compared to the nominal resolution and efficiency of the detector. We focus on noble element targets, as energy sharing with an extended molecule will result in a systematically broadened energy peak. These measurements would provide a valuable test of the precision of the theoretical calculations upon which multiple branches of physics rely.

In this paper we outline a general purpose CRES detector capable of detecting X-rays with high energy resolution. We present a description of the apparatus in Section II and an exploration of the anticipated principles and interactions that inform the detailed setpoints of the detector in Section III. Then in Section IV we apply the principles to a system optimized for measuring the atomic energy levels of argon and its photoelectric cross-section with 6-keV X-rays. Section IV also contains...
a description of the spectral deconvolution required to reconstruct the original X-ray spectrum. We end with a discussion of considerations beyond the example argon system.

II. APPARATUS

To build an X-ray detector using CRES principles, we rely on photoelectric interactions between the X-ray and a target atom. The target is embedded in a Tesla-scale magnetic field that causes ejected electrons to undergo cyclotron motion, emitting microwave radiation. Magnetic trapping coils then keep the electron confined to an instrumented volume, allowing for long-term collection of the radiated microwaves as shown in Fig. 1. The signal from the microwave antenna is amplified and digitized, and a Fourier transform applied to create a power spectrum for any given time segment.

Consecutive maxima in the power spectra can be correlated to create a track that represents the energy of the electron. The resulting plot shows both a gradual energy loss to the cyclotron radiation as well as discrete energy changes stemming from shallow-angle scatters with the target atoms. Fig. 2 shows an example spectrogram from Asner et al., \[13\]. Reconstruction of the initial frequency of the electron can be transformed into an ejected-electron energy via the equation

\[
f = \frac{eB}{2\pi (m_e + T/c^2)} \tag{1}\]

where \(e\) is the charge on the electron, \(B\) is the magnetic field, \(T\) is the electron’s kinetic energy, \(c\) is the speed of light, and \(m_e\) is the mass of the electron.

From Eq. (1) we see that the resolution of the energy reconstruction is related to the accuracy with which we measure the starting frequency of the track, and there are a number of parameters which contribute to this accuracy. In the case of a constant radiofrequency (RF) signal, the resolution of the measurement is inversely proportional to the number of samples recorded before the Fourier transform is performed; therefore, the longer the measurement time, the finer the frequency resolution. Since the frequency of the radiating electron is changing, however, there is a limit to this approach, because applying the Fourier transform to a changing signal will result in a broad feature in the resulting power spectrum. Another effect is the length of the track, where too short a track will preclude a high-quality fit to the starting point. A third effect is the power of the signal in the first bin, because if the power happens to fluctuate below the noise level, the measured starting frequency will be systematically high. Further exploration of these effects on energy resolution are discussed in Section III.

The maximum cyclotron frequency occurs at the lowest energy, and for an electron in a 1-T field, this ends up being approximately 27.99 GHz, while for a 30-keV electron the cyclotron frequency is 26.44 GHz. A typical digitizer operates at 1 GS/s requiring the signal to be downmixed to below the Nyquist frequency of 500-MHz to achieve a suitable sample rate. This is why the frequencies plotted in Fig. 2 have been reduced by 24 GHz in hardware. Multiple stages of downmixing were utilized in Ref. \[13\], and we encourage readers to review that work for additional details.

The readout chain has a bandwidth of a few hundred MHz, requiring the experimenter to tune the mix down to capture signals for the desired electron energy. To achieve a wider effective bandwidth, the signal would need to be amplified and split into multiple mix-down chains, each with the local oscillator tuned to a different frequency. An instrumented range of 23.4-27.9 GHz enabled by parallel downmixers would be able to simultaneously record electrons between 1 and 100 keV while retaining single-eV energy precision throughout.

As a final comment on the apparatus design, we note that, depending on the configuration of the decay chamber, the recorded signal can have systematic effects that can lead to misidentification of the main microwave carrier signal \[10\], leading to an error in the energy reconstruction. These effects must be accounted for in the analysis stage.

III. SYSTEM PERFORMANCE

In this section we discuss optimization of the experimental setup. We review choices of target ma-

![Schematic of CRES system optimized for X-ray detection](image)
terial and density, the effect of electron trap depth, and considerations of event rate and pileup rejection.

A sparse noble gas is an ideal target material for a CRES X-ray detector, as it meets several requirements. The target material must have high purity so that electronegative molecules do not absorb the trapped electron, and noble gasses are relatively straightforward to recirculate and purify. Also, because noble elements do not combine into stable molecules, no molecular shifts in the energy spectrum must be considered when deconvolving the raw spectrum (see Section IV).

The target gas density must be optimized to find a balance between sufficient interaction targets and mean free path between electron scatterers. If the electron mean free path is very short, the tracks visible in Fig. 2 will be too short to reconstruct the starting frequency. Although the tracks visible in that image are on the order of 1 ms, improved algorithms have been shown to reliably reconstruct tracks down to 200 µs [17]. To obtain a 1/e time between scatterings of 200 µs, it behooves us to set the target density according to the total electron interaction cross-section for any given target material. Using the Evaluated Electron Data Library [18], we summed the elastic, large-angle elastic, ionization, bremsstrahlung, and excitation cross-sections as a function of electron energy for all the stable noble elements, and used a logarithmic interpolation between available datapoints. Not all individual interaction cross-sections were tabulated down to 1 keV, and a linear extrapolation was used to extend all curves to this value, as it proved more stable than a logarithmic extrapolation. The results are shown in Fig. 3.

The total interaction cross-section can be used to calculate the mean free path, and thereby the mean time between scatters, of the trapped electron as a function of both electron energy and target medium. If further improvements in event reconstruction can accurately determine starting frequencies with tracks shorter than 200 µs, the density of the target can be increased to enhance the data rate.

Higher-Z elements will generally have larger photoelectric interaction cross-sections than lighter elements, but at the cost of greater complexity in the spectral analysis (see Section IV). The photoelectric cross-section for the stable noble elements is shown in Fig. 4. At energies near 6 keV, the photoelectric interaction rate will be maximized with a xenon target medium, given an isobaric constraint. Note, however, that the argon-electron interaction cross-section is smaller than that of xenon-electron interactions, allowing for a greater target density for a given free time between scatters. Because the photoelectric cross-section at 6 keV is approximately equal for argon and krypton, an optimized argon system will have an overall higher event rate than an optimized krypton system at this energy.

Next we consider the effect of the depth of the electron trap. Electrons emitted perpendicular to the main magnetic field (θ = 90°) will spiral in place with no axial motion, while electrons emitted parallel to the magnetic field (θ = 0°) are guaranteed to escape the trap. For small trapping fields, Esfahani
et al. 2019 give the trapping angle as

$$\theta_{\text{trap}} = \sin^{-1} \left( \sqrt{1 - \frac{B_{\text{trap}}}{B_{\text{main}}}} \right)$$

(2)

The higher the trapping field, the more electrons will be trapped. There is a point of diminishing returns, however, related to the power emitted by the spiraling electron, given by

$$P = \frac{\pi}{\epsilon_0} \frac{2e^2}{3c} f_0^2 \beta^2 \sin^2 \theta \frac{1}{1 - \beta^2}$$

(3)

where $f_0$ is the cyclotron frequency as the electron kinetic energy goes to 0, and $\beta = v/c$. The power radiated is maximal when $\theta = 90^\circ$, and vanishingly small when the electron is emitted parallel to the main B field. It is possible that an electron, though trapped within the active volume according to Eq. (2), nonetheless results in no observable signal because the radiated power is too small relative to the system noise to reconstruct the tracks in the power spectrum. In this situation, the performance of the system would not be improved by increasing the trapping field unless the electronics and radiofrequency noise were reduced to increase the statistical significance of the lower-power signal.

Next we consider the bias in electron trajectory following a photoelectric interaction. Fig. 5 shows the distribution in the emission angle of the electron with respect to the main magnetic field, assuming the incident X-rays are parallel to said field. A trap depth of 95 mT on a 1 T main field will trap electrons between 72° and 108°. At the extremes of this range, the power radiated by a trapped electron will be at 90% of maximum, and therefore still likely to be properly reconstructed, relative to an electron emitted at 90°. In this range, an incoming 1-keV photon will have a 42% probability of ejecting a trapped electron. At 1 MeV, the forward momentum of the ejected electron population reduces that probability to about 2%. It may be possible that, if an experiment is optimized to detect photons greater than 100 keV, placing the source to the side of the active volume, rather than off one end, may increase the overall detection efficiency.

Related to the electron trajectory is the effect of wall collisions. If an electron that would otherwise be trapped is emitted within two cyclotron radii of a physical surface, the electron will impact that surface before a full cyclotron revolution. Thus the effective radius of the system is reduced by two cyclotron radii. For large systems, the remaining volume will be sufficient to perform a measurement in a timely manner, but there is a lower bound on the radius of the system of approximately 2.5 mm, below which the electron is almost guaranteed to be absorbed, precluding observation of any RF signal.

Finally we turn our attention to issues of pileup. Given the very sparse target density (see Section IV), the rate of simultaneous events is anticipated to be very low. If, however, two photons were to interact at exactly the same time, there are several parameters that can be used to separate the events:

- Energy, where two simultaneous ejected electrons will result in two distinct tracks separated in frequency on the power spectrum, rather than as a single track at the summed energy
TABLE I. X-ray energies and relative intensities from the decay of $^{55}$Fe [20].

| Energy (eV) | Relative Intensity |
|------------|---------------------|
| 5887.65    | 51                  |
| 5898.75    | 100                 |
| 6490.45    | 20.5 (combined)     |
| 6535.2     |                     |

TABLE II. Electron binding energies for argon, from Ref. [3].

| Shell | $K$ | $L_1$ | $L_2$ | $L_3$ | $M_1$ | $M_2$ | $M_3$ |
|-------|-----|-------|-------|-------|-------|-------|-------|
| Energy (eV) | 3206 | 326   | 251   | 248   | 29.3  | 15.9  | 15.7  |

IV. EXAMPLE CASE: MEASUREMENTS OF ARGON TARGET WITH 6-KEV X-RAYS

We now work through an example of an experiment to measure the argon atomic energy levels and photoelectric cross-section from low-energy X-rays from the decay of $^{55}$Fe. This source emits photons at energies shown in Table I while Table II shows the binding energies of electron shell levels in the argon atom. For the last two energies in Table II near 6500 eV, the value of 20.5 is the combined relative intensity. For simplicity, and given the ambiguity of this tabulation, in the following analysis we assume the full 20.5 relative intensity is entirely in the 6490.45-eV X-rays, with no X-rays emitted at 6535.2 eV.

Electrons ejected via a photoelectric interaction will have the energy of the incident X-ray minus the binding energy of the shell from which the electron was ejected. In Fig. 6 we show the expected energy spectrum for $^{55}$Fe on an Ar target, taking into account the photoelectric cross-sections given by Scofield. For this spectrum, we have assumed a constant energy resolution of 1 eV across all energies.

The most intense electrons in this scenario are near 2700 eV, with falling intensities up to 6500 eV. This drop in intensity derives primarily from falling photoelectric cross sections on the outer electron shells, although the lower intensity of the 6.5-keV X-rays relative to the 5.9-keV X-rays also contributes to lower electron intensity in the upper energy region. To optimize the system for the lowest-energy electrons (i.e., those with the greatest interaction cross-section according to Fig. 3), we use the total electron interaction cross-section at 2.7 keV to determine the target density. The cross-section for argon at this energy is $2.06 \times 10^6$ barns/atom. A 2.7-keV electron has a velocity of $30.7 \times 10^5$ m/s, and if we require a 1/e time between scatters of 200 $\mu$s, the mean free path must be 6.14 km. This path length can be achieved by setting the target density to $7.91 \times 10^9$ atoms/cm$^3$, or 2.23 $\times 10^{-7}$ Torr. At higher electron energies the cross-section decreases, so the track length of those electrons will be longer and therefore more efficiently reconstructed.

To calculate the event rate, we will assume the active volume is 10 cm long and 3 cm in diameter. We used a Monte Carlo approach to calculate the distribution of path lengths in the active volume, where the source was modeled as an extended planar disk 1 cm in diameter, coaxial with the active volume, 1 cm away from a flat face, and with isotropic emission. Averaging over all trajectories, including those that did not intersect the active volume, resulted in an average path length of 0.39 cm.

A more full-featured Monte Carlo calculation would be necessary to incorporate additional physical effects of a real-world design, such as attenuation through a beryllium window for an external source.
where the variable definitions and their values are shown in Table III. We use the average path length in the active volume in Eq. (1) as a simplification, which is valid given the very long (∼82,000 km) attenuation length of the X-rays in the target medium. We have assumed that the trap depth is shallow enough that any trapped electrons will radiate sufficient power to be seen above noise levels, and hence we set $\epsilon_{pwr}$ to full efficiency (see Section III).

The final rate of detected K-shell ejections from the 5899 eV X-rays, i.e., counts in the peak at 2693 eV, is 0.275 Hz. We would collect approximately 166,000 events per week into this peak, with approximately half that into the peak at 2682 eV. The L-shell peaks and the K-shell ejections from the 6.5 keV X-rays would have 10,000 to 20,000 counts in this same time frame. The lowest-intensity peaks would have approximately 120 counts. With a bin width of 0.5 eV and a total systematic resolution of 1 eV, this lowest-intensity peak would have a centroid uncertainty of approximately 0.1 eV. This accuracy would allow measurement of the relative energy levels to better than an eV, while the number of counts in each peak would provide a relative measurement of the photoelectric cross-sections, with accuracy ranging from 0.2% to 10%.

The spectrum shown in Fig. 6 assumes a constant $\epsilon_{eff}$ across all electron energies. In reality, because the argon-electron cross-section drops at higher energies, $\epsilon_{eff}$ would be higher, and the number of counts in these peaks correspondingly higher. When performing a full spectral analysis not only of the peak energies, but intensities, this effects must be incorporated. Otherwise, the greater number of counts in these higher-energy peaks would be incorrectly interpreted as a higher photoelectric cross-section.

The cyclotron frequency of electrons between 2500 and 6500 eV is 27.64 to 27.86 GHz for a 1 T field. Thus if 27.44 GHz is subtracted from the signal using a downmixer, and the system’s bandwidth was at least 215 MHz, the full spectrum could be recorded with frequencies between 200 and 415 MHz.

Once a spectrum is obtained, it must be deconvolved to reconstruct the original X-ray emissions. The lowest-energy peak in the spectrum needs to be associated with higher-energy peaks from any L- and M-shell ejections from the same energy of incoming photon. Establishing this correlation will increase the significance of the primary peak. The lowest-energy peak should then be augmented by the binding energy of the K-shell electron, and normalized by the photoelectric cross-section to recover the original X-ray energy and intensity. The next-lowest unassociated peak will then be identified, with its own associated L- and M-shell correlations, and augmented in the same manner as the first peak. Each peak will therefore be identified in turn. Thus the full spectrum of the original X-ray energies and intensities is reconstructed. The heavier target atoms have more shells from which electrons can be ejected, leading to a more complicated spectrum with a greater number of associated peaks, although the analysis approach presented here is still applicable.

We end this section with a concept for a systematic test, where the $^{55}$Fe source is replaced by a $^{49}$V source. This source has a comparable half-life, and emitted X-ray spectrum to the $^{55}$Fe, and provided the argon target properties are consistent, differences in the resulting spectra can be attributed directly to the use of a different source. A custom radioactive source might incorporate both $^{55}$Fe and $^{49}$V, removing many systematic differences between data acquisition runs.

![Table III](image)

TABLE III. Variables used in calculating the event rate on an argon target with an $^{55}$Fe source. $L$ comes from a Monte Carlo calculation of the unattenuated path length through the active volume, $\sigma$ comes from Scofield [5], $\epsilon_{emi}$ comes from Section III, and $\epsilon_{tr}$ is set by establishing the 1/e track length attenuation constant to 200 $\mu$s. The efficiency for detecting an electron up to $\pm18^\circ$ away from perpendicular, $\epsilon_{pwr}$ is assumed to be 100%.
measurement comes from both the instrumental resolution and the precision of the incoming photons. Given the energy and precision constraints, a good choice for irradiation usable for all the noble targets might be the 60 keV gammas from the decay of $^{241}\text{Am}$.

We now consider the event rate between the lightest and heaviest stable noble elements to delineate the extremes of system performance. The photoelectric cross-section at 60 keV is seven orders of magnitude greater for xenon than helium. The electron interaction cross-section, though, is two orders of magnitude greater. This simplistic consideration would give an event rate in the xenon $10^5$ times that of the event rate in the helium, but with a big caveat: the population of the M- and N-shell peaks from the xenon are also attenuated by several orders of magnitude, in a manner similar to what is shown for the L and M shells for argon in Fig. 6, while the helium target only has a single shell to characterize. A full design consideration of the two targets and an $^{241}\text{Am}$ source might show a surprising parity between the required run times.

As previously mentioned, the system can be optimized for any particular X-ray energy and target medium. The optimization process starts with determining the shortest acceptable path length of the trapped electron, based on the total interaction cross-sections in Fig. 3. This path length then determines the target density. Given the very long attenuation length of the sparse targets, the event rate will scale directly with the volume, although the observation of the CRES signal in a large volume will require alternative readout hardware, such as that described for the RF Demonstrator for Phase III of Project 8 [21].

As a final concept, this approach to high-precision X-ray spectroscopy can be expanded to characterization of external materials. If the source in Fig. 1 is replaced with a beryllium window, a target outside that window can be induced to emit X-rays via irradiation with photons (e.g., from $^{241}\text{Am}$, similar to measurements performed by Turhan et al. [22]), or electrons from a sufficiently intense electron gun. Once the energy levels of the gas in the active volume are determined to high precision, the energy levels of external targets can also be measured and potentially used for identification of unknown samples.

VI. SUMMARY

In this work we have presented a concept for measuring the energy levels and photoelectric cross-sections for the shells of the stable noble elements using cyclotron radiation emission spectroscopy. The apparatus is anticipated to have eV-scale resolution, thereby testing the theoretical calculations underpinning many physics experiments to unprecedented accuracy. We have described experimental considerations to optimize a target, with dependences on photon energy, target density, photoelectric cross-section, electron interaction cross-section, electron emission angle distribution, active volume and source configuration, electron trapping depth, and signal in the power spectrum above the noise floor. With high-resolution measurements of the noble element energy levels this work can be extended to characterize external materials if the internal source were replaced with a low-Z window and the external material is illuminated by photons or electrons of a compatible energy.

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