A simple conversion electron detector for Mössbauer source experiments

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A vibration-free conversion electron detector has been constructed for Mössbauer source experiments with extremely low doping levels ($\sim$1 ppm) of cobalt-57. While the overall efficiency is somewhat less than that of microfoil systems, the design is greatly simplified and it still yields more than a sixfold enhancement in signal-to-noise ratio over standard transmission methods. The lightweight detector ($\sim$25 g) may be mounted directly on a conventional transducer without limiting the performance. This allows low-temperature source measurements to be made by moving the detector and fixing the sample/source in a cryostat. The overall linewidth (HWHM) of the detector is 0.16 mm/s with the broadening being due to saturation effects in the enriched foil.

I. INTRODUCTION

Mössbauer measurements are made mainly in transmission due to the simplicity of the experimental arrangement and the large counting rates. However, when working with low f factor transitions, analyzing radiation either emitted from low activity sources or scattered from samples (e.g., in Rayleigh scattering or selective excitation double Mössbauer), the signal-to-noise ratio is limited primarily by the detector/analyzer efficiency. The optimum solution is a conversion electron detector using microfoils (typically 300 nm thick);[1] however, construction of such devices is complicated by the fragility of the foils. The design presented here retains most of the advantages of microfoil detectors but uses only commerically available parts, greatly simplifying manufacture and reducing cost.

Following resonant absorption of the 14.4 keV γ ray, only about 10% of the ^{57}Fe nuclei return to the ground state by reemitting a γ ray, the rest emit either x rays or conversion electrons,[2] almost two electrons being emitted, on average, for each resonant absorption event. The relatively low-energy electrons ($\sim$7 keV) are generally detected by constructing a proportional counter around the sample: the (grounded) sample forms the cathode and one or more fine wires biased to $\sim$1 kV form the anode plane. The usual fill gas is helium, to minimize sensitivity to γ rays, with a few percent methane added as a quench gas. Conversion electron Mössbauer spectroscopy[3] has been widely used in nondestructive testing and surface analysis and may be modified to yield a high efficiency detection system. In principle, only resonant events are detected (nonresonant photons simply pass through the system), leading to a great enhancement of signal-to-noise ratio compared to conventional transmission methods. However, two factors limit performance: (i) the escape depth of the conversion electrons is small ($\sim$100 nm) compared to the penetration depth of the 14.4 keV γ ray ($\sim$13 μm in iron) meaning that only a small fraction of the resonant material in a normal sample is actually used; (ii) all of the incident radiation can generate photoelectrons from the sampling leading to a nonresonant background rate.

Efficiency may be maximized by making the resonant foils so thin that essentially all of the conversion electrons produced escape to be detected. A sufficient number of such foils and anode planes may be placed in sequence so as to fully absorb the incident radiation. This device is a microfoil conversion electron (MICE) detector.[1,4] For ^{57}Fe the optimum foil thickness is about 300 nm. Stainless steel is generally used as it gives a single line absorber and it is usually prepared from iron enriched to about 90% in ^{57}Fe in order to maximize the resonant cross section and minimize the amount of nonresonant material in the detector. Two such foils are required. Despite the technical difficulties associated with their construction, such detectors have been used and greatly enhance the signal-to-noise ratio, yielding a factor of 10 increase in efficiency over the conventional proportional counter and absorber arrangement.[1] Recently, a lightweight design was reported which can be mounted on a velocity transducer.[5]

Most of the difficulties in constructing and operating a MICE detector are associated with the extremely thin foils. As they are not commerically available they must be rolled from thicker stock; moreover, they are fragile once made. Great care must be taken in mounting the foils in order to minimize vibrations which will otherwise cause line broadening. These problems led us to investigate a simpler design. In the present paper we describe a vibration-free version of a simple conversion electron detector which combines the simple configuration of a normal conversion electron detector and the high signal-to-noise ratio of a MICE detector. The detector can easily be constructed from commerically available components and has a mass of less than 25 g allowing it to be driven in constant-acceleration mode by a conventional Mössbauer drive, without limiting velocities.

II. DETECTOR DESCRIPTION

Figure 1 shows a photograph of the detector mounted on the Mössbauer drive. The cross section of the detector is

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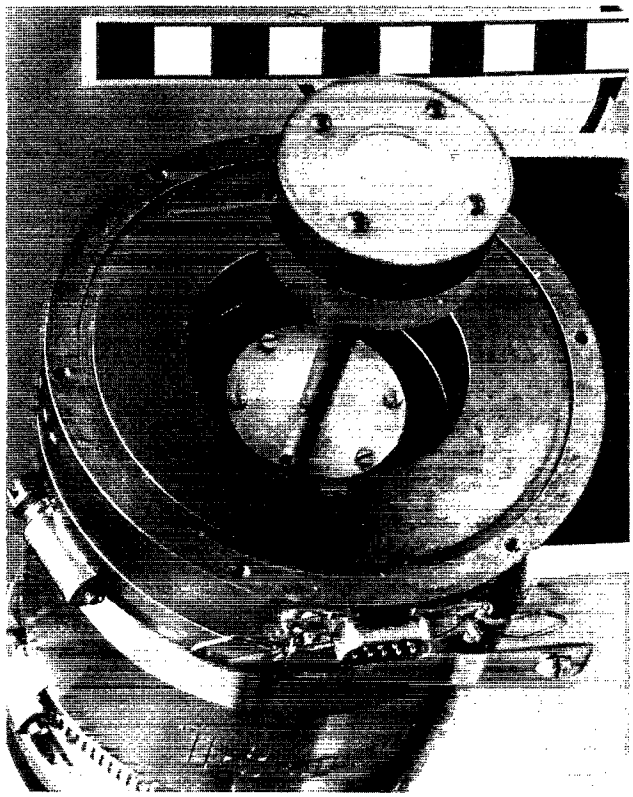

FIG. 1. Photograph of the conversion electron detector mounted on the Mössbauer drive. A centimeter scale is shown at the right.

shown in Fig. 2. The detector body, made of Plexiglas, consists of three parts: (i) front window, (ii) anode wire housing, and (iii) back plate. The front window is a 0.12 mm aluminized Mylar sheet epoxied to a Plexiglas plate 40 mm in diameter, 2 mm thick with a 20-mm-diam aperture in the center. The 25 μm gold-coated tungsten anode wire is supported in the center of the detector by 1.4 mm o.d. copper capillaries epoxied into holes drilled in the detector

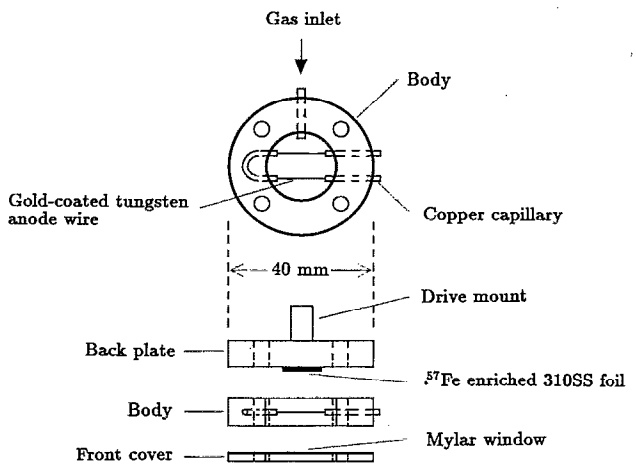

FIG. 2. Sectional views of the detector. Top diagram illustrates a section through the anode plane and the bottom one shows a side view of the separated components.

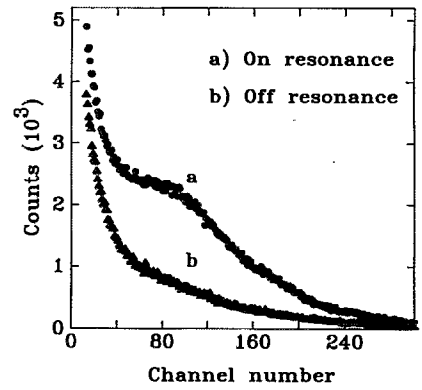

FIG. 3. Pulse height spectrum obtained using a 2 m Ci source with the conversion electron detector (a) on resonance and (b) off resonance.

body. These tubes are equidistant from the central axis and are 5 mm apart. The ends of the anode wires are soldered to the tubes. This anode configuration provides a large active detector volume. A 1.3 μm 310 stainless-steel foil 90% enriched in ^{57}Fe (Ref. 6) is cemented to the aluminized rear side of the detector with a conducting adhesive which keeps the foil firmly fixed and maintains good electrical contact with the grounded inner body of the detector. Facing surfaces were polished so as to achieve good contact but not a hermetic seal. Finally, all three parts are held together with four brass bolts. The interior space is a 20-mm-diam cylinder 8 mm in height. All inner surfaces of the detector are fully aluminized and connected to ground. A 250-mm-long, 2.5 mm o.d. lightweight coaxial cable (RG-174/u) is used between the detector and preamplifier to minimize mechanical stiffness which would interfere with driving the detector. The gas enters through a 2 mm o.d. plastic tube, and exits between the joints in the detector parts. A constant gas flow is used to prevent air leaking back into the active region. All possible measures have been taken in order to reduce the mass of the detector, which could affect the linearity of the acceleration when the detector is mounted on the drive. The present detector has a total mass of less than 25 g.

The detector is operated as a conventional continuous-flow gas-filled proportional counter using premixed helium/4% methane at a flow rate of 40 cm^3/min (chosen to optimize counting efficiency). Too low a flow rate leads to air leaking into the detector resulting in a significant loss of efficiency. With more attention to sealing a lower flow rate could certainly be used, and complete sealing combined with a dedicated exit port would certainly yield more reproducible performance.[1] The operating voltage was determined by observing the amplifier output and counting rate as the bias voltage was increased from 800 to 1200 V in 50 V steps. In our case, the operating voltage was chosen to be 1050 V. Lower voltages yield reduced gain, making operation difficult, while some breakdown was observed at higher voltages. Some background contributions are always present. One due to detector noise and low-energy photoelectrons can be reduced by optimizing the lower level discriminator setting (about channel 30 on Fig. 3 was

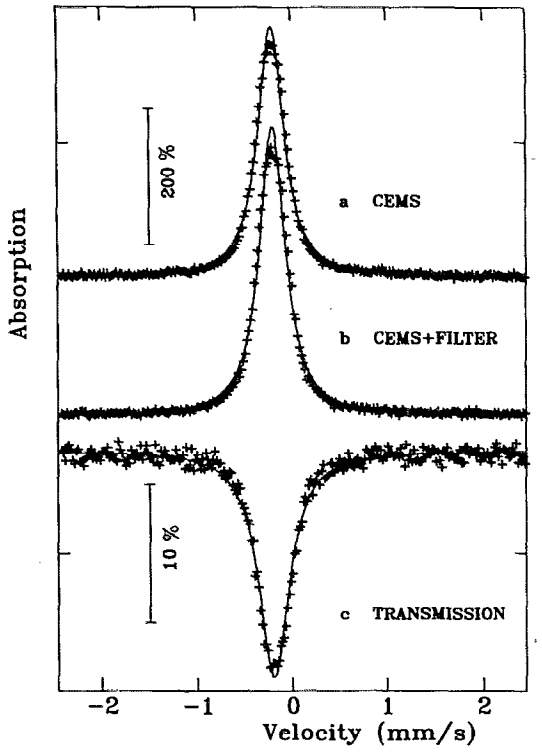

FIG. 4. Mössbauer spectra of 310 stainless steel measured using the conversion electron detector (CED) (a) without and (b) with a 1.7 mm plexiglass filter in front of the source to absorb the Fe K_α x rays. (c) shows a comparison spectrum measured in transmission using an 8 μm 310 stainless-steel foil. The substantial improvement in the signal-to-noise ratio of the CED spectra is apparent. All spectra were measured in the same geometry for the same time.

used). Another, due to photoelectrons produced by incident 6.5 keV Fe K_α x rays from the source can be reduced using a 1.7 mm Plexiglas filter in front of the source, although at the expense of a slightly reduced count rate. The remaining background of photoelectrons comes from nonresonant 14.4 keV photons and higher-energy photons (122 and 136 keV) from the source which are scattered by nonresonant materials (e.g., in the stainless steel, detector body and wires) in the detector. This latter contribution is minimized by using low-Z materials as fast as possible.[7]

III. DETECTOR PERFORMANCE

Figure 3 shows the pulse height spectrum obtained with 2 m Ci ^{57}CoRh source on and off resonance. Background events were minimized by setting the lower level discriminator to approximately channel 30. In Fig. 4 we compare the Mössbauer spectrum obtained using the conversion electron detector with a transmission spectrum obtained for the same counting time and geometry using an 8 μm 310 stainless-steel absorber prepared from natural iron. For fixed counting time and geometry, we obtained more than a sixfold increase in signal-to-noise ratio using the conversion electron detector compared with the standard transmission measurement, and the linewidth (HWHM) is also reduced: 0.16(1) mm/s compared with 0.19(1) mm/s from the transmission spectrum. This latter change is primarily due to the reduction of saturation effects. The performance of the detector is summarized in Table I. We note that the primary source of improvement in this detector design is reduction of background at the expense of some loss of total signal strength. Microfoil systems which allow signal detection from both faces of several foils yield a little more than double the counting efficiency with a further suppression of background counts as a result of the smaller total mass of foils in the beam. Even though our simplified design allows detection of electrons from only a single face of the foil, we still obtain over a factor of six enhancement in signal-to-noise ratio over the transmission measurement, indicating that much of the complexity of MICE systems can be avoided for ^{57}Fe measurements.

TABLE I. Summary of conversion electron detector (CED) performance compared with standard transmission measurement (TRANS) on an 8 μm 310 stainless-steel foil. All spectra were recorded for the same time and in identical geometry. The numbers in brackets are the fraction of incident photons leading to the signal; in the case of the CED background the fraction is overestimated by a factor of 10 as the contributions of the 122 and 136 keV γs are not included in the incident beam measurement.

	Baseline (counts)	Signal (counts)	Linewidth (mm/s)	Effect (%)	Signal/Noise
Incident beam	27732	...	...	...	...
CED	459 (1.7%)	3082 (11%)	0.16	671	144
TRANS	21904 (79%)	3312 (12%)	0.19	15	22

IV. EXAMPLE OF APPLICATION

We are currently studying cobalt substitutional site preferences in a range of iron-based intermetallic compounds.[8–10] To do this we incorporate about 1 ppm of cobalt as ^{57}Co (the radioactive parent of the 14.4 keV ^{57}Fe transition) by coreduction of $^{57}CoCl_2$ and $^{56}Fe_2O_3$ followed by appropriate alloying. ^{56}Fe is used to eliminate resonant self-absorption in the source which would otherwise prevent any signal being observed. The extremely low doping level means that the hyperfine parameters associated with the emission spectrum are essentially unchanged from the reference (cobalt-free) sample thus allowing unambiguous site assignments. Furthermore, since the active material is incorporated as cobalt, but decays to iron *before* emitting the γ ray used in the Mössbauer experiment we obtain the normal iron subspectra weighted according to the site occupation of the cobalt. The subspectra intensities therefore provide a direct measure of the cobalt site statistics. Thickness restrictions on the sample, necessary to minimize nonresonant self-absorption, mean that activities of $\sim$0.4 m Ci are generally used.

As a specific example of the application of the detector we show data on the compound Nd_2Fe_{17} (Ref. 10) which has the rhombohedral Th_2Zn_{17} structure with four iron sites: 18f, 18h, 9d, 6c (referred to the hexagonal unit cell). Since Nd_2Fe_{17} has a Curie temperature of 330 K, the iron sites of the spectrum are not resolved at room temperature [Fig. 5(a)]; however, the spectrum obtained at 90 K, by

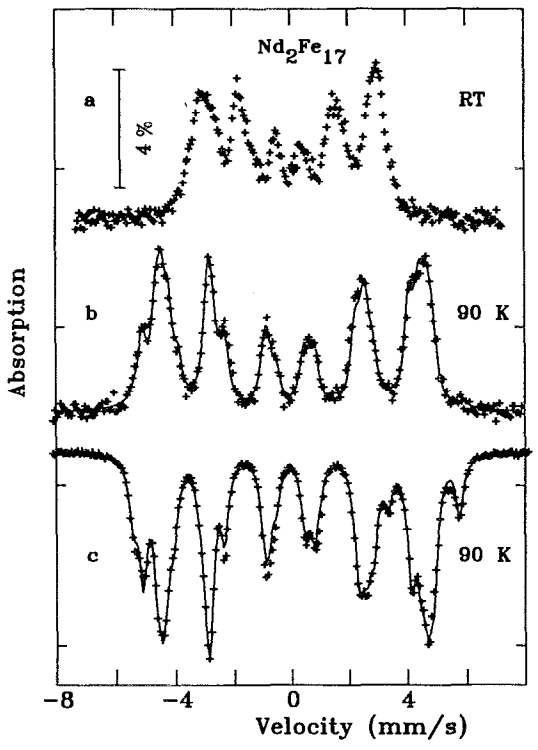

FIG. 5. Mössbauer spectra of cobalt-doped Nd_2Fe_{17} obtained using the conversion electron detector (a) room temperature and (b) 90 K. The transmission spectrum of the reference material at 90 K is shown in (c). Note the absence from (b) of the line at +5.3 mm/s showing that cobalt avoids occupying the site associated with that line.

moving the detector with the sample/source placed in a conventional cryostat, is clearly resolved and subspectra of the individual sites can be distinguished [Fig. 5(b)]. If the cobalt simply substituted randomly for iron in the structure, then the source spectrum would be the same as that of the reference (cobalt-free) material shown in Fig. 5(c), however, a visual comparison reveals that this is not the case. It is thus clear that cobalt substitutes preferentially on certain sites. The most obvious difference is the line at +5.8 mm/s (due to the 6*c* site) which is absent from the source spectrum, and shows that cobalt almost completely avoids this site. More accurate computer analysis further shows that cobalt also avoids the 18*f* site while preferentially occupying the 18*h* and 9*d* sites.

V. CONCLUSIONS

We have constructed a vibration-free conversion electron detector for Mössbauer source experiments with extremely low doping levels of ^{57}Co. Instead of sequential microfoils, we have used a single, much thicker, foil fixed in the detector. The detector weighs less than 25 g allowing measurements to be made at low temperature by moving the detector and placing the sample/source in a cryostat. Moreover, fixing the resonant foil to the back plate of the detector eliminates the possibility of vibrational broadening of the measured spectrum. The advantages of the present design over the MICE detector are ease of construction with lower cost and weight. While the counting efficiency is somewhat lower than that of a MICE detector, our design yields a comparable signal-to-noise ratio, and represents a sixfold enhancement over standard absorption methods.

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