Feasibility study on the micro-foil internal conversion electron (MICE) detector application to high-temperature emission Mössbauer spectroscopy

A. Błachowski and K. Ruebenbauer

Mössbauer Spectroscopy Division, Institute of Physics, Pedagogical University
PL-30-084 Kraków, ul. Podchorążych 2, POLAND

(a) Corresponding author: sfrueben@cyf-kr.edu.pl
Tel.: +(48-12) 662-6317, 19
Fax: +(48-12) 637-2243

Short title: MICE in high-temperature Mössbauer spectroscopy

Abstract

Detailed calculations have been performed to assess potential applicability of the MICE-CEMS detector (CEMS - conversion electron Mössbauer spectroscopy) to the high temperature emission Mössbauer spectroscopy by means of the 14.41keV transition in $^{57}$Fe. The comparison was made with the standard transmission method using either proportional or semiconductor detector. It was found that for typical source matrices used in the investigations performed at high temperatures the MICE-CEMS detector yields poorer results than the high quality semiconductor detector. Therefore the development of the dedicated MICE-CEMS detector is unjustified for this particular application.

Keywords: high temperature, MICE, emission Mössbauer spectroscopy

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1. Introduction

The Mössbauer spectroscopy in the energy domain [1] is very useful tool to look upon microscopic mechanisms of diffusion [2-4]. In order to study behaviour of extremely diluted impurities one has to resort to the emission Mössbauer spectroscopy [5-7]. In such case one is going to investigate Mössbauer level formed in the source due to the decay of the parent nucleus. Sources in the form of single crystals yield the most complete information [2]. Usually thin sources are used including thickness for the resonant self-absorption [7], i.e., sources highly transparent to the emitted radiation including recoilless photons. Investigations are generally performed at high temperatures and under specific thermodynamic conditions, e.g. under controlled atmosphere surrounding the sample under investigation [7]. In most cases the emission line is composed of the single, albeit broadened Lorentzian line, and the emitted radiation can be considered as unpolarised. One has to bear in mind that the recoilless fraction is significantly reduced at elevated temperature [8]. The best Mössbauer line for the purpose is the 14.41 keV line resulting from the single photon radiative decay of the first excited level of the stable nucleus of $^{57}$Fe. The beam coming out of the source has to be reasonably collimated in order to observe desired direction in the single crystal. On the other hand, strong sources are hard to prepare and maintain under the above mentioned conditions. The best parent available for the line mentioned is $^{57}$Co decaying via the electron capture [8]. The lifetime of this parent is long enough to consider it as a stable source during the single data accumulation time period.

Basically two types of resonant detectors are possible. One can use some single line unpolarised resonant absorber in combination with the high energy resolution X-ray detector having high efficiency for the Mössbauer line and low efficiency for the background lines, the latter having high energies. Some low atomic number filter is usually inserted in front of the X-ray detector to attenuate intensity of the fluorescent X-ray lines originating from the Mössbauer atom in the source and resonant absorber. Geometry has to be designed in such a way to approximate closely true transmission conditions. The exit windows of the sample chamber and X-ray detector are usually made of beryllium. The beryllium has to be free of the resonant atoms, of course. It is very difficult to apply Doppler motion [9] to the source, as the source is located in some chamber under unusual thermodynamic conditions. Hence, one is moving the resonant absorber in front of the above X-ray detector, the latter being equipped with the X-ray filter. The best linearity of motion is available in such case as one can apply high quality Mössbauer transducer in this arrangement [10]. The best single line absorbers for the line in question can be made of the random substitutional and polycrystalline alloy of highly enriched in $^{57}$Fe iron in natural rhodium. The absorber is maintained usually at room temperature. The practical enrichment factor is about 95 at.%, while one can use the alloy containing up to about 20 at.% of iron in the vicinity of the room temperature without measurable broadening due to the difference in the hyperfine interactions for various resonant atoms in the absorber. The atomic resonant cross-section is practically Lorentzian for the line considered as the transition is almost pure M1 transition [8]. Therefore its amplitude is orthogonal to the amplitude of the E1 transition, the latter amplitude being a dominant contribution to the photo effect and Rayleigh scattering on the resonant atom at the energy of the Mössbauer transition [11-13]. Usually there are no problems with the X-ray detector saturation effects of any kind as the available beams are rather weak. Hence, solutions reducing background by means of some Bragg scattering are out of reach in the present context – particularly due to the fact that the beam is far from being perfectly collimated [14]. A collimation sufficient for the studies of the diffusive motions and sufficient to avoid spread of velocities is still unsatisfactory to achieve reasonable efficiency of the Bragg scattering.
Another possible approach is to use internal conversion electron resonant detector called CEMS - (conversion electron Mössbauer spectroscopy) [15]. A transition considered here is strongly converted and therefore such approach has to be taken into account. Detectors of the remaining secondary radiation are obviously less appealing as they are strongly susceptible to the unavoidable background. A detector has to be attachable to the transducer for the reasons outlined above, and hence resonant targets are to be rigid enough to withstand vibrations without producing line broadening. The resonant part of the detector has to be made from the same alloy as used in the transmission arrangement in order to get as narrow absorption cross-section as possible. The main point is that the range of the conversion electrons within such material is very small compared to the typical absorption length even for the highly enriched targets. Mullen et al. [16] proposed to circumvent this problem by using multiple thin targets MICE (micro-foil internal conversion electron detector). In such case the best solution is to use gas type detector working in the proportional regime and filled with the low atomic number gas to assure insensitivity to the X-ray radiation [16]. Rigidity of targets can be achieved by deposition of the resonant layers on the low atomic number substrates free of the resonant atoms. Metallic material for resonant layers and substrates is the best choice in order to avoid local charging of the detector interior.

The aim of the present contribution is to estimate feasibility of such MICE detector in conjunction with the high temperature emission Mössbauer spectroscopy. A comparison with the standard transmission arrangement is made as well. The paper is organised as follows: section 2 is devoted to the theoretical background and approximations used in this work, while section 3 shows results of some simulations based on the previous formalism. The last section 4 is devoted to the conclusions.

2. Theoretical background

A resonant part of the emission Mössbauer line can be described in the present context by the following expression [17]:

\[ \rho(\omega) = \frac{\Gamma_s}{2\pi} \frac{f_s}{(\Gamma_s / 2)^2 + (\omega - \omega_0)^2}. \]  

(1)

Here the symbol \( \Gamma_s \) denotes half width of the emitted line, where the following condition is satisfied \( \Gamma_s \geq \Gamma > 0 \) with \( \Gamma \) being the natural half width (to very good approximation). The symbol \( 0 < f_s < 1 \) stands for the recoilless fraction of the source in the direction pointing from the source towards the resonant detector. Finally, \( \omega \) stands for the ambient energy of the emitted radiation (usually in the first order Doppler velocity units), while the symbol \( \omega_0 \) stands to good approximation for the resonant energy of the source in the same units. The beam falls perpendicular on the resonant layer. Therefore transmission to the depth \( x \geq 0 \) can be described by the following equations [17]:
\[ T(v, \omega, x) = T_\rho(v, \omega, x) + T_\sigma(x) + \sum_{j=1}^{J} T_{\phi}^{(j)}(x), \]

\[ T_\rho(v, \omega, x) = \rho(\omega) \exp \left[ -n_0 \sigma_0 f_\sigma x \left( \frac{\Gamma}{\Gamma_\sigma} \right) \frac{(\Gamma_\sigma/2)^2 + (\omega - \omega_0 - \delta \omega - \nu)^2}{(\Gamma_\sigma/2)^2 + (\omega - \omega_0 - \delta \omega - \nu)^2} \right] \exp[-n \sigma x], \]

\[ T_\sigma(x) = (1 - f_\sigma) \exp[-n \sigma x] , \]

\[ T_{\phi}^{(j)}(x) = A_j \exp[-n \sigma_{\phi}^{(j)} x]. \]

(2)

Here the symbol \( v \) stands for the applied Doppler shift along the beam, \( n_0 \) denotes the concentration of the resonant nuclei within the target, \( \sigma_0 \) stands for the resonant cross-section for absorption, and the symbol \( 0 < f_\sigma < 1 \) denotes recoilless fraction of the target. The symbol \( \Gamma_\sigma \geq \Gamma \) stands for the resonant half width in the target, and \( \delta \omega \) denotes a total shift, the latter observed between the source and the target. The symbol \( n \) denotes concentration of all atoms in the target and \( \sigma \) stands for the effective non-resonant atomic cross-section for absorption of the photons belonging to the Mössbauer line (either recoilless or with recoil) in the target. The symbol \( \sigma_{\phi}^{(j)} \) denotes corresponding non-resonant cross-section for absorption of other photons belonging to the background radiation line having the \( j \)-th index and originating within the source. Subsequently the symbol \( A_j \geq 0 \) stands for the ratio of the background intensity due to the \( j \)-th background line to the total intensity of the resonant line (including recoilless and with recoil intensities). The term \( T_\phi(v, \omega, x) \) corresponds to the resonant radiation, \( T_\sigma(x) \) describes radiation emitted with the creation or annihilation of some phonons, while the term \( \sum_{j=1}^{J} T_{\phi}^{(j)}(x) \) is due to the background radiation. The symbol \( J \geq 1 \) denotes the number of background lines involved. Usually one can neglect other background than originating in the source including background of the detector and electronics itself. Secondary radiation (mainly originating in the detector) can be neglected as well under normal circumstances except internal conversion electrons due to the decay of the resonant nuclear level. Electrons generated directly in the interactions of the incoming photons with the atomic shells of the resonant target are to be taken into account as well - practically those are electrons generated by the photo effect. Properly designed detector is unlikely to see other scattering mechanisms, the Compton scattering being dominant of the latter.

Therefore a thin layer having thickness \( dx > 0 \) becomes a source of the energetic electrons. In our case one can consider each resonant atom as independent of other atoms as the interference effects are cancelled due to the random character of the alloy. Particular contributions to the electron source can be expressed as follows:

\[ dS(v, x) = dS_{\rho}^{(a)}(v, x) + dS_{\rho}^{(b)}(v, x) + dS_{\sigma}(x) + \sum_{j=1}^{J} dS_{\phi}^{(j)}(x), \]

\[ dS_{\rho}^{(a)}(v, x) = \int_{-\infty}^{+\infty} d\omega \left( \frac{\alpha}{1 + \alpha} \right) n_0 \sigma_0 f_\sigma \left( \frac{\Gamma}{\Gamma_\sigma} \right) \left( \frac{(\Gamma_\sigma/2)^2 + (\omega - \omega_0 - \delta \omega - \nu)^2}{(\Gamma_\sigma/2)^2 + (\omega - \omega_0 - \delta \omega - \nu)^2} \right) T_\rho(v, \omega, x) \ dx, \]

\[ dS_{\rho}^{(b)}(v, x) = \int_{-\infty}^{+\infty} d\omega \ n_0 \sigma T_\rho(v, \omega, x) \ dx, \]

\[ dS_{\sigma}(x) = n_0 \sigma T_\sigma(x) \ dx, \]

\[ dS_{\phi}^{(j)}(v, x) = n_0 \sigma_{\phi}^{(j)} T_{\phi}^{(j)}(x) \ dx, \]

\[ dS_{\phi}^{(j)}(x) = n_0 \sigma_{\phi}^{(j)} T_{\phi}^{(j)}(x) \ dx. \]

(3)
Here the symbol $\alpha > 0$ stands for the total internal conversion coefficient of the resonant atom in the target. A contribution $dS_{u}^{(s)}(v,x)$ is due to the conversion electrons following resonant absorption. A contribution $dS_{b}^{(b)}(v,x)$ is due to the electrons generated by the resonant radiation in the non-resonant processes. On the other hand, contributions $dS_{n}(x)$ and $\sum_{j\neq 1} dS_{b}^{(j)}(x)$ are due to non-resonant processes caused by Mössbauer photons emitted with recoil and background radiation, respectively.

One has to bear in mind that the attenuation of the X-ray beam is mainly due to the resonant absorption and photo effect at the relevant photon energies. On the other hand, electrons of the energy involved here enter diffusion regime almost immediately after creation. Hence, the energy resolution of the electron spectrum is practically lost (one has to open acceptance window of the single channel analyser as widely as practical) except for extremely thin targets, the latter being impractical here. Therefore one can use the effective range for particular electron groups, while attempting to calculate the probability for the particular electron to emerge from the target into the working gas of the detector. One can further assume that all electrons emerging from the target give signal (pulse) in the detector, the latter being accepted by the data storage system. In such case simple geometrical considerations make possible to calculate the above probability provided the target thickness exceeds the maximum range of the electrons (see inset of Fig. 1). We accept this assumption, as CEMS targets are always too thin from the point of view of the incoming X-ray radiation. Hence, the signal of the detector can be calculated as follows:

$$
S(v) = \frac{1}{2} \left[ \int_{0}^{R_{e}} dS_{u}^{(s)}(v,x) \left( 1 - \frac{x}{R_{e}} \right) + \int_{0}^{R} dS_{b}^{(b)}(v,x) + dS_{n}(x) \left( 1 - \frac{x}{R} \right) + \sum_{j\neq 1} dS_{b}^{(j)}(x) \left( 1 - \frac{x}{R_{b}^{(j)}} \right) \right].
$$

(4)

The factor $\frac{1}{2}$ is due to the fact that resonant targets are deposited on the substrate, the latter being almost completely transparent to the X-ray radiation, but completely opaque to the electrons. The symbols $R_{e} > 0$, $R > 0$ and $R_{b}^{(j)} > 0$ denote effective ranges of the conversion electrons, electrons generated by the Mössbauer line (either recoilless or with recoil) in the non-resonant processes and background radiation due to the particular background line, respectively. Usually one can neglect signal (non-resonant) generated in other parts of the detector. The above expression applies to the back-scattered electrons.

Hence, one can conclude that the resonant target is to be made of the iron free beryllium sheet covered both sides by the resonant layer – or single side for the first and last sheet. Some very thin rhodium interface has to be laid between beryllium and the resonant layer. A resonant layer has to be covered on the side exposed to the working gas by similar interface covered additionally by the extremely thin protective layer of gold separating rhodium from the working gas (usually natural helium with several mol.% of iso-pentane at about $\frac{1}{3}$ of the ambient pressure). A single side target has to be covered by the similar gold layer on the non-resonant side. The working medium has to be kept under static conditions as the detector is to be attached to the transducer. Very thin iron free tungsten wires can be used as anodes (about 20 $\mu$m diameter). Possible design of such detector is shown in Fig. 1. It is important to prepare carefully substrate surface prior to deposition of the above layers in order to maintain uniformity of the resonant layer.
Figure 1.
Typical layout of the MICE detector applicable to the high temperature emission Mössbauer spectroscopy is shown. Inset shows arrangement of the resonant layers and diffusion range of electrons (in a schematic way).

The above formalism can be easily extended to the MICE type detector provided all resonant layers have the same properties as usually expected. The MICE signal can be expressed as follows upon having performed integration over the target thickness:

\[
S_{\text{MICE}}(v) = P(v) + S_0 , \quad \text{where:}
\]

\[
P(v) = \frac{1}{\omega} \int_{-\infty}^{\infty} d\omega \rho(\omega) \sum_{k=1}^{K} \exp \left\{ -\left[ L(\omega,v) + n\sigma \right] (k-1) d \right\} G_i(k)
\]

\[
\left\{ \left. \frac{\alpha}{1+\alpha} \right| L(\omega,v) F_i(\omega,v,R_n) + n\sigma F_i(\omega,v,R) \right\} \quad \text{and}
\]

\[
S_0 = \frac{1}{\omega} \sum_{k=1}^{K} \left\{ \int_{-\infty}^{\infty} \sum_{j=1}^{J} A_j \cdot G_i(k,j) \exp \left[ -n\sigma^{(j)} (k-1) d \right] F_j(\omega^{(j)},\sigma^{(j)}) \right\} .
\]

Here the index \( K \geq 1 \) stands for the number of resonant layers employed, while the symbol \( d > 0 \) denotes thickness of each resonant layer, the latter being greater than the maximum range of the electrons. The abbreviation \( L(\omega,v) \) stands for the following function:

\[
L(\omega,v) = n_0 \sigma_0 f_a \left( \frac{\Gamma}{\Gamma_a} \right) \left( \frac{(\Gamma_a/2)^2}{(\Gamma_a/2)^2 + (\omega - \omega_0 - \delta\omega - v)^2} \right).
\]
Remaining abbreviations have the following form for the back-scattering resonant layers:

\[
F_1(\omega, v, R) = \frac{1}{L(\omega, v) + n\sigma} \left( 1 - \frac{1}{R[L(\omega, v) + n\sigma]} \left( 1 - e^{-\frac{d[L(\omega, v) + n\sigma]}{R}} \right) \right),
\]

\[
F_2(R, \sigma) = 1 - \frac{1}{n\sigma R} (1 - e^{-\alpha R}).
\]  

(7)

Similar functions for the forward scattering layers, i.e., for the electrons emerging along the incoming radiation beam take on the form:

\[
F_1(\omega, v, R) = \frac{1}{R[L(\omega, v) + n\sigma]} e^{-\frac{d[L(\omega, v) + n\sigma]}{R}} \left( 1 + \left[ L(\omega, v) + n\sigma \right] R \right),
\]

\[
F_2(R, \sigma) = \frac{1}{n\sigma R} e^{-\alpha R} \left[ e^{\alpha R} - (1 + n\sigma R) \right].
\]  

(8)

A detector with single resonant layer usually operates in the back-scattering mode. Detectors with multiple layers should have even number of the resonant layers. The first one (closest to the source) has to operate in the forward scattering mode (see Fig. 1). One can assume as well that other parts of the detector than the resonant layers and the back wall practically do not interact with the incoming beam of radiation for properly designed detector. The absorption of the remainder of the beam in the back wall of the detector (see Fig. 1) has no effect on the signal in the case of proper design. One can account eventually for the beam attenuation by multiple substrates setting functions \( G_1(k) \) and \( G_2(k) \) in the expression (5). For a detector with single layer these functions equal unity. Otherwise they are to be defined as follows:

\[
G_1(k) = \exp[-n_s\sigma_{S0}D_s f(k)] , \quad G_2(k, j) = \exp[-n_s\sigma_{S0}^{(j)}D_s f(k)].
\]  

(9)

Here the symbol \( n_s \) stands for the concentration of atoms in the substrate, \( \sigma_{S0} \) denotes cross-section for the absorption of the Mössbauer photons (recoilless and with recoil), while the symbol \( \sigma_{S0}^{(j)} \) stands for the corresponding cross-section for the background radiation, the latter being due to the \( j-th \) background line. The symbol \( D_s > 0 \) stands for the thickness of the substrate (all substrates should have the same thickness). The function \( f(k) \) takes on the form \( f(k) = \frac{k}{2} \) for back-scattering resonant layers, and the form \( f(k) = \text{int}(\frac{k}{2}) + 1 \) for the layers scattering forward. It is assumed that electrons generated in the substrate are unable to penetrate resonant layer and to get into the working gas. For resonant layers thinner than respective electron range, corresponding upper limits in the integrals of the expression (4) are to be replaced by the resonant target thickness \( d \). In such cases some electrons generated in the substrate are able to penetrate into the working gas of the detector. However they can be neglected for substrates having very low atomic numbers as for the latter case respective cross-sections for the photon absorption are very small. The electron scattering is inefficient in such substrates as well, and hence back-scattered electrons penetrating into the working gas can be neglected too. One has to note that functions \( F_2(R, \sigma) \) described by expressions (7) and (8) transform under such circumstances as follows: 


One has to redefine a bit standard transmission integral expression in order to make a comparison with the traditional transmission arrangement [18]. Namely, the signal in the transmission mode can be expressed as follows:

\[
1 - \left( \frac{1}{n\sigma R} \right) \left( 1 - e^{-n\sigma R} \right) \Rightarrow 1 - e^{-n\sigma d} \left[ 1 - \left( \frac{1}{n\sigma R} \right) \left( 1 - e^{-n\sigma d} (1 + n\sigma d) \right) \right],
\]

\[
\left( \frac{e^{-n\sigma d}}{n\sigma R} \right) \left[ e^{n\sigma R} - (1 + n\sigma R) \right] \Rightarrow 1 + \left( \frac{1}{n\sigma R} \right) - \left( \frac{d}{R} \right) - e^{-n\sigma d} \left( 1 + \frac{1}{n\sigma R} \right).
\]

(10)

Here the symbol \( 10 \leq \varepsilon < 1 \) stands for the X-ray detector efficiency for photons in the vicinity of the resonant line, and \( D > 0 \) stands for the thickness of the resonant absorber used in the transmission arrangement. It is assumed that the resonant absorber is made of the same material as the CEMS resonant targets, and that it remains under the same conditions. The parameter \( \lambda \geq 1 \) stands for the ratio of the total count rate within the single channel analyser window to the count rate due to the Mössbauer line within this window [18]. The last count rate is a sum of the count rate due to the photons emitted with and without recoil. A standard assumption is made that the X-ray detector and single channel analyser are tuned to the photo peak of the Mössbauer line. Re-emitted resonant line from the resonant absorber (either recoilless or with recoil) can be eliminated from the detector entrance by proper design of the geometry.

A comparison is possible under assumption that the number of photons originating in the Mössbauer nuclear transition in the source and falling on the MICE detector is the same as the number of photons falling on the resonant absorber and accepted by the single channel analyser in the transmission mode. Usually one can assume that the count rate far off resonance remains constant as either MICE detector or absorber execute motion of the very small amplitude, the latter being performed around stable average position. The loss of counts (if any) during data processing can be made the same for both methods. One has to assume as well that geometry is the same in both cases. In such case one can multiply respective signals described by the equations (5) and (11) by the scaling factor \( LN / 0 \), where \( 0 \gg N \) stands for the number of photons described above. The symbol \( L \) stands for the number of the final data channels and it is assumed to be the same for both modes. The number of data channels has to be large enough to avoid histogram effects of various kinds. All channels have the same width on the velocity and time scales. Resulting data can be further scattered applying standard statistical scatter by means of the Monte-Carlo method in order to get some results for visual comparison. The above scatter is the dominant source of errors for properly designed arrangement. One can assume that the dispersion in each data channel equals square root of the channel content in the large statistics limit applicable here. A distribution around the expected average value has Gaussian shape in each channel under the above assumption.

Another simple criterion can be applied to unscattered data provided the velocity range is broad enough to encompass significant portion of the far off resonance signal at low and high velocities both. Namely, one can calculate relative error of the observed effect according to the equation:
Here the symbol \( Y_0 \) stands for the maximum signal in the CEMS mode and minimum of the signal in the TRANS (transmission) mode. On the other hand, \( Y_i \) and \( Y_L \) represent signals in the first and the last data channels, respectively. The above data are obtained scaling respective expressions, i.e., (5) and (11) by the factor \( N_0/L \). A quality factor can be defined as \( Q = \beta(\text{CEMS})/\beta(\text{TRANS}) \). The application of MICE detector is justified provided the following condition is satisfied: \( Q < 1 \). Otherwise one has to resort to simpler transmission mode. A difference in the count rates due to the X-ray filter effect on the Mössbauer line could be accounted for adjusting the parameter \( \varepsilon \). This filter might act slightly different in the CEMS mode in comparison with the TRANS mode. The beam falling onto respective resonant detectors has to be sufficiently collimated to avoid spread of the velocities.

One can expect that with the raising temperature of the source all parameters remain constant except \( f_r \), \( \Gamma_r \) and \( \delta \omega \). The last parameter changes mainly due to the second order Doppler shift varying in the source with the varying source temperature. Its variation has no effect on the data quality provided the velocity range is sufficiently broad. On the other hand, remaining two parameters have effect on the maximum of the resonant flux of photons, the latter defined by the equation (1). Namely, this maximum is equal to \( (2f_r)/(\pi\Gamma_r) \). Hence, it is interesting to investigate variation of the quality factor \( Q \) versus changes in \( f_r \) and \( \Gamma_r \). The last parameter \( (2f_r)/(\pi\Gamma_r) \) drops down with the increase of the temperature.

The above theoretical background is approximate, of course, as many minor effects have been neglected. However, it seems complete enough to draw some solid conclusions about the MICE detector potential in application to the emission Mössbauer spectroscopy in the high temperature limit. Microphonic effects due to the anode vibrations during normal operation of the detector does not seem a serious problem as the detector cannot be treated as truly spectrometric. Some processes leading to the energy absorption within the detector are likely to produce multiple electrons, the latter electrons emerging from the targets to the working gas area. However these events produce single pulses in the detector due to the limited time resolution of the detector (about 1\( \mu \)s).

\[
\beta = \sqrt{Y_0 + \frac{1}{2}(Y_i + Y_L)} \left| Y_0 - \frac{1}{2}(Y_i + Y_L) \right|.
\]

(12)

2.1. Calculation of the electron range

Electrons loose energy due to the two dominant processes, i.e., due to the ionisation and radiative scattering (Bremsstrahlung). The latter process can be neglected here as the electrons have insufficient energy to make this process significant even for targets having large atomic numbers. Therefore other mechanisms can be neglected as well, as they require even higher energies to play any role. The loss of the kinetic energy \( E \) due to ionisation can be expressed in the following way in accordance with the Bethe-Bloch formalism [19]:

\[
\frac{dE}{dx} = \sigma_s \left( m_e c^2 \right)^2 \rho \langle Z \rangle \left[ \ln[\beta(E)] \left( \frac{E + m_e c^2}{\beta^2(E)} \right) \left( \frac{E}{m_e c^2} \right)^{1/2} - \frac{1}{2} \beta^2(E) \right].
\]

(13)
Here the symbol $\sigma_e = 10^{-24}\text{ cm}^2$ stands for the cross-section for the electron scattering on another electron, the latter cross-section is treated in the classical limit. The symbol $m_e c^2$ stands for the rest energy of the free electron (the symbol $m_0$ denotes rest mass of the free electron, and the symbol $c$ stands for the velocity of light in vacuum). On the other hand, the symbol $\rho$ denotes the concentration of the scattering atoms. The latter atoms have the following weighed average atomic number $\langle Z \rangle$. The symbol $\beta(E)$ denotes the following expression:

$$\beta(E) = \frac{\sqrt{E(E + 2m_e c^2)}}{E + m_e c^2}.$$  \hspace{1cm} (14)

The average ionisation potential energy (effective) can be expressed as follows [19] in accordance with the Bethe-Bloch formalism corrected for the electron scattering in the low energy range:

$$I(E) = a \langle Z \rangle + b \langle Z \rangle \left( \frac{m_e c^2}{E} \right)^{3/2}.$$  \hspace{1cm} (15)

Here the symbols $a > 0$ and $b > 0$ represent constants. A particular range $R_{j}^{(j)}$ can be obtained as a solution of the following equation:

$$\varepsilon_0 = E^{(j)} - E_j + \int_0^{R_{j}^{(j)}} \left( \frac{dE}{dx} \right) dx.$$  \hspace{1cm} (16)

The symbol $\varepsilon_0 > 0$ denotes the exit potential energy of the electron (average work function), the latter leaving target. The symbol $E^{(j)}$ denotes the energy of the $j-th$ radiation line (including resonant line - either recoilless or with recoil), while the symbol $E_j$ stands for the internal binding energy of the primary electron released due to the absorption of the radiation. The average range due to the $j-th$ radiation line takes on the form:

$$R^{(j)} = \sum_j C^{(j)}_j R^{(j)}_j \text{ with } \sum_j C^{(j)}_j = 1.$$  \hspace{1cm} (17)

Weights $C^{(j)}_j$ represent relative contributions due to various atomic shells contributing to the production of either internal photo or conversion electrons.

### 3. Results of simulations

Somewhat simplified decay scheme of the $^{57}\text{Co}$ atom is shown in Fig. 2. Some marginal branches of the decay are neglected. The creation of the electron holes in other shells than the K shell of the resonant atom is neglected as well, as long as the radiation source is considered. It is further assumed that all of the above shells are filled by the emission of the Fe – Kₐ photon – in the source of the radiation. The above simplifications are justified, as other processes are really marginal [20]. Basically one has to consider five lines: 136 keV line (1),
122 keV line (2.), 14 keV line – resonant (3.), Fe – Kα line (4.) and eventual Kα fluorescent line of the source matrix provided the matrix is mono atomic (5.). Intensities of the above lines are modified by the finite source thickness and by the non-resonant radiation filter inserted in front of the detector. The above intensities normalised by the total intensity of the resonant line can be expressed as follows for a uniform source – at the entrance of the detector:

\[
I_1 = b_R \left[ \frac{1 + \alpha(3)}{1 + \alpha(1)} \right] \exp \{ [\mu(3) - \mu(1)]d_F \} A_1(d_s),
\]

\[
I_2 = \left[ \frac{1 + \alpha(3)}{1 + \alpha(2)} \right] \exp \{ [\mu(3) - \mu(2)]d_F \} A_2(d_s),
\]

\[
I_3 = 1,
\]

\[
I_4 = b_R \left[ \frac{\alpha(1)[1 + \alpha(3)]}{1 + \alpha(1)} \right] + \left[ \frac{\alpha(2)[1 + \alpha(3)]}{1 + \alpha(2)} \right] + \alpha(3) \exp \{ [\mu(3) - \mu(4)]d_F \} A_4(d_s),
\]

\[
I_5 = C \exp \{ [\mu(3) - \mu(5)]d_F \}. \tag{18}
\]

Here the symbol \( b_R \) stands for the branching ratio of the direct transition from the 136 keV level to the transition over the cascade. Symbols \( \alpha(l) \) denote respective total conversion coefficients, while the symbols \( \mu(l) \) denote corresponding attenuation coefficients due to the presence of the filter. The symbol \( d_F \) stands for the thickness of the filter, while the index \( l \) takes on the following values \( l = 1, 2, 3, 4, 5 \). The parameter \( C \) denotes the relative intensity of the fluorescent line due to the matrix, as seen on the surface of the source exposed to the detector. The functions \( A_i(d_s) \) take on the following form:

\[
A_i(d_s) = \left( \frac{\mu_s^{(3)}}{\mu_s^{(l)}} \right) \frac{1 - \exp[-\mu_s^{(l)}d_s]}{1 - \exp[-\mu_s^{(3)}d_s]}.
\]

\[
(19)
\]
Here symbols $\mu_i^{(i)}$ denote respective attenuation coefficients, while the symbol $d_s$ stands for the source thickness. It has to be noted that the 136 keV transition has negligible recoilless fraction in the vicinity of the room temperature or at higher temperatures. Splitting of the electron shells due to the fine interactions can be neglected here. The use of the total conversion coefficient $\alpha(3)$ roughly compensates for the neglected emission of the respective Auger electrons.

It is sufficient to take into account photoelectrons generated at the deepest available atomic levels in the resonant targets. On the other hand, the conversion electrons originating from the iron K, L and M shells within the resonant target are to be taken into account. All electrons generated due to the E2 transition can be neglected together with other electrons following decay of the resonant nuclear state. The fine structure of the atomic levels can be neglected in this case, too.

All parameters used to calculate the spectral shape at the entrance of the detector for two types of the source matrix [8,20,21] are summarised in Table I. On the other hand, all relevant electron ranges within the resonant target are listed in Table II [19,22]. The last column of this table contains all ranges used in further simulations. Parameters of the resonant target substrates are listed in Table III [21]. Remaining constant parameters used in simulations are listed in Table IV [8].

Simulations were performed varying the source width $\Gamma_s$, the source recoilless fraction $f_s$, changing the number of resonant layers $K$ and thickness of the resonant layer $d$. Two source matrices were investigated, i.e., Rh and CoO. A comparison was made with the Kr-filled proportional detector and with the intrinsic Si detector. A linear velocity range covering $\pm 3$ mm/s was applied in all cases except for the largest source width $\Gamma_s$, where the range $\pm 6$ mm/s was applied. Resulting quality factors $Q$ are summarised in Table V. A detector composed of ten resonant layers having thickness 75 nm each was chosen for further investigations. The quality factor was calculated varying source recoilless fraction and source width for both matrices and detectors used in the transmission geometry. Results are shown in Fig. 3. On the other hand, spectra simulated for the CEMS detector made of ten layers having 75 nm thickness each are plotted in Fig. 4 and compared with the corresponding spectra obtained for the intrinsic Si detector. The first pair of spectra was calculated for the Rh source matrix with the following parameters: $\Gamma_s = 0.10$ mm/s and $f_s = 0.78$, while the second pair of spectra was calculated for the CoO source matrix with the parameters: $\Gamma_s = 0.75$ mm/s and $f_s = 0.22$. The latter parameters correspond to the CoO source temperature of about 1450 K and to the relatively high oxygen pressure of the atmosphere surrounding the source [7]. Fluorescent X-ray lines originating in the oxygen atoms of the CoO source were neglected as they have insufficient energies to emerge either from the source or to penetrate the window of the chamber containing the source. Other fluorescent lines could be neglected for properly designed source holder as well.
Figure 3.
The quality factor is shown versus source recoilless fraction and for various source widths. Horizontal dashed lines show the border between transmission and CEMS regions. Symbols along right vertical axes denote the following source widths: 1 - 0.1; 2 - 0.2; 3 - 0.5; 4 - 1.0 and 5 - 2.0 mm/s.

Table I

Source and radiation parameters used in simulations. The branching ratio was adopted as $b_R = 0.875$, and two source matrices have been considered: Rh with $d_s = 10\, \mu m$ and CoO with $d_s = 40\, \mu m$. The non-stoichiometry of the latter matrix has been neglected, as it is small for all available thermodynamic conditions [7]. It has been assumed that the radiation filter is made of graphite having $d_F = 1\, mm$ and kept at ambient temperature. The parameter $C = 0.15$ of the equation (18) was adopted for both sources. A resonant line is transmitted to the detector entrance with the following efficiency 0.66 and 0.59, respectively.

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<th>Energy [keV]</th>
<th>$\alpha(l)$</th>
<th>$\mu(l)$ [cm$^{-1}$] graphite</th>
<th>$\mu_{s1}(l)$ [cm$^{-1}$] Rh</th>
<th>$\mu_{s1}(l)$ [cm$^{-1}$] CoO</th>
<th>$I_f$ Rh</th>
<th>$I_f$ CoO</th>
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<tr>
<td>Fe - $K_\alpha$</td>
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<td>4400.37</td>
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Figure 4. Mössbauer spectra simulated for the CEMS detector and compared with the transmission spectra collected by means of the intrinsic Si detector having 2 mm thickness.

Table II
Electron range in the resonant target composed of 80 at.% of Rh and 20 at.% of Fe. The weighed atomic number equals $\langle Z \rangle = 41.2$, the average atomic density equals $\rho = 7.45 \times 10^{22} \, \text{cm}^{-3}$, while the following values for the remaining parameters have been adopted: $a = 11 \, \text{eV}$, $b = 0.15 \, \text{eV}$ and $e_o = 4.3 \, \text{eV}$. The last parameter is the exit potential energy (average work function) from the polycrystalline gold protective layer. Other conversion coefficients than listed below were neglected due to their smallness [20]. Average range stands for the weighed average range according to the equation (17).

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Table III
Parameters of the Be substrate having thickness $d_s = 0.2$ mm and atomic density $n_s = 1.23 \times 10^{23}$ cm$^{-3}$. Cross-sections for photo effect are shown [21].

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<th>Cross-section [barn/atom]</th>
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<td>Rh - $K_\alpha$</td>
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<td>Fe - $K_\alpha$</td>
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Table IV
Remaining constants used in simulations. Parameters $\lambda$ and $\varepsilon$ have been set as 1.2 and 0.6 for the Kr-filled proportional detector, and as 1.02 and 0.994 for the intrinsic Si detector having 2 mm thickness. The parameter $N_0$ was set to $2 \times 10^4$, while the number of data channels was set to $L = 2047$, the latter being available using, e.g. the MsAa-3 spectrometers. The total shift $\delta\omega$ was set arbitrarily to zero, as it is irrelevant in the present context.

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<th>Comments</th>
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<td>$\alpha$</td>
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<td>total conversion coefficient of the resonant line</td>
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Table V
Quality factor $Q$ calculated versus number of layers and layer thickness. The recoilless fraction $\gamma$ and the source width $\Gamma$ were set as 0.78 and 0.10 mm/s for Rh matrix and as 0.22 and 0.75 mm/s for CoO matrix, respectively. Bold frames mark optimum MICE detectors still technically feasible.

**Rh source matrix**

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**CoO source matrix**

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4. Conclusions
The CoO matrix is obviously closer to the expected matrices to be investigated at high temperatures. On the other hand, the signal to noise ratio is somewhat overestimated for either the transmission method or for the electron conversion method. The re-emission of the
resonant line toward the detector is neglected in the former case, while the Compton background is neglected in the latter case. The quality factor is defined as the ratio, and therefore it is insensitive to the above over estimations - in the first order. It is clear that the quality factor is unfavourable for the MICE detector particularly when compared with the high quality semiconductor detector. Therefore one can conclude that the development of the MICE detector for the high temperature emission spectroscopy utilising 14.41 keV transition in $^{57}$Fe is unjustified.

The situation is different for extremely strong beams of radiation (here beams are rather weak), where the limiting factor is mainly due to the saturation of the detector instead of the count rate as encountered here. In such case detector efficiency becomes irrelevant and one can push the signal to the background ratio to the extreme high limit. Hence, the noise due to the background can be reduced to almost null. However extremely thin targets are required to achieve this goal and therefore single layer detectors are to be used instead of the MICE detectors. In principle one can approach the following limit

$$ R_{SB} = \frac{\sigma \epsilon_f f_a \alpha}{\sigma (1 + \alpha)} $$

of the signal to the background ratio for the detector similar to the one considered above. Here the symbol $\sigma$ denotes the cross-section for the photo effect on the particular atomic shell of the resonant atom, while the symbol $\alpha$ denotes corresponding internal conversion coefficient for the resonant radiation. For the values of the parameters adopted in this paper and for the K-shell of $^{57}$Fe one obtains roughly $R_{SB} = 2.4 \times 10^4$% provided $f_s = f_a = 0.78$. One needs almost as thin layer of the resonant atoms as the mono-layer without almost any support and some electron spectrometer of the high (energy) resolution in order to get close to this limit. The incoming beam has to be cleaned by some medium resolution Bragg monochromator. One can further increase the above $R_{SB}$ factor provided the incoming beam is linearly polarised. Namely, one can arrange the scattering geometry in such a way to avoid photoelectrons on the entrance to the electron spectrometer. This is possible as the amplitude for the photo effect is practically of the E1 character, while the amplitude for the resonant absorption is practically of the pure M1 character for the nuclear transition in question. These two amplitudes are mutually orthogonal. Hence, in principle one can remove background completely. It is obvious that the efficiency of such arrangement is extremely low. Therefore it is unlikely to improve the data quality unless the original source is extremely strong. In conclusion, one cannot use as the almost sole criterion the above $R_{SB}$ factor as such approach is likely to be simply wrong.

References