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OBSERVATION OF A DOPPLER BROADENING OF THE 4438 keV GAMMA-LINE OF ^{12}C IN PROCESSES $^{12}C(n, n'\gamma)^{12}C$ AND $^{9}Be(\alpha, n\gamma)^{12}C$

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A Doppler broadening is described of the 4438 keV spectral gamma-line observed by means of a Ge(Li) detector during the deexcitation of nuclei of ^{12}C in an inelastic scattering $^{12}C(n, n'\gamma)^{12}C$ using an ^{241}Am–Be source as well as during the reaction $^{9}Be(\alpha, n\gamma)^{12}C$ taking place in the Am–Be source. The FWHM of the spectral line is equal to (90 ± 4) keV in the latter reaction and (64 ± 8) keV in the former process. Experimental values agree well with theoretical ones.

Introduction

We studied the Doppler broadening of the 4438 keV spectral gamma-line, monitored by a Ge(Li) detector during the deexcitation of nuclei of carbon ^{12}C accompanying the inelastic scattering $^{12}C(n, n'\gamma)^{12}C$ and the reaction $^{9}Be(\alpha, n\gamma)^{12}C$. The study of this kinematic effect was prompted by the results of experiments concerned with the determination of carbon content in various materials by the method of inelastic neutron scattering using a radionuclide neutron source ^{241}Am–Be.

Theory

In nuclear processes of the type $(n, n'\gamma)$, $(\alpha, n\gamma)$, $(p, \alpha\gamma)$, $(n, \alpha\gamma)$ and others the resulting nucleus is in an excited state and is then deexcited by emitting gamma-radiation. If these processes take place on light target nuclei the resulting

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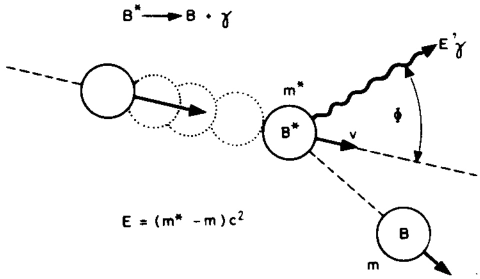

Fig. 1. Decay of the excited nucleus during flight with the emission of gamma-radiation

nuclei after the interaction possess usually a considerable kinetic energy and their deexcitation may take place already during the flight. This type of deexcitation requires that the mean lifetime of the resulting excited nucleus should be shorter than its time of flight through the medium. In this case the energy of the photon E'_γ of the gamma-radiation will differ from the photon energy E_γ emitted in the nucleus at rest. The phenomenon is called a Doppler energy shift. If the excited nucleus emits a photon in a direction at an angle Φ relative to the direction of flight of the nucleus (Fig. 1), the energy of the photon will be defined by the formula:

$$E'_\gamma = \frac{1}{2} \cdot \frac{E(E + 2\,mc^2)}{E + mc^2} \sqrt{1 - \left(\frac{v}{c}\right)^2} \cdot \frac{1}{1 - \frac{v}{c} \cos \Phi}, \tag{1}$$

where E – transition energy,
m – mass of nuclei,
v, c – velocities of the nucleus in the medium and of the light in a vacuum, respectively.

Eq. (1) follows from the kinematics of the nuclear disintegration during a flight and, provided $v/c \ll 1$ and $E/mc^2 \ll 1$, it may be modified to the well-known formula

$$E'_\gamma \cong E_\gamma \left(1 + \frac{v}{c} \cos \Phi\right) \tag{2}$$

where

$$E_\gamma = E\left(1 - \frac{E}{2\,mc^2}\right) \tag{3}$$

is the energy of photon emitted by a nucleus at rest. Eq. (2) implies that the energy shift does not take place at $\Phi = 90°$, in the range of angles of $0° \leqslant \Phi < 90°$ the energy $E'_\gamma > E_\gamma$ and at $90° < \Phi \leqslant 180°$ the energy $E'_\gamma < E_\gamma$ and the Doppler energy shift thus takes place.

A real case does not involve the disintegration of a single excited nucleus but a whole set of excited nuclei moving in different directions and at different velocities. The shift of energy of the photons of the gamma-radiation in individual events in the emission process leads in this case generally to the shift and broadening of the spectral line peak. This broadening is called the Doppler broadening; its magnitude depends on the difference between the maximum and minimum observed energy of photons for the given transition. In other words, the broadening of peak corresponding to the spectral line depends on the minimum and maximum velocities of the moving excited nuclei. These limit velocities of the nuclei may be calculated from the kinematics of the process which gives rise to excited nuclei disintegrating further through the emission of gamma-photons.[1] The shape of the spectral line, i.e. the energy distribution of the observed photons of the given transition, depends on the geometrical arrangement of the experiment and on the position of the detector (observer).

We observed a broadening of the 4438 keV line corresponding to ^{12}C. The source of the excited nuclei of $^{12}C^*$ was the reaction $^9Be(\alpha, n)^{12}C^*$ and the inelastic scattering $^{12}C(n, n')^{12}C^*$. The reaction $^9Be(\alpha, n)^{12}C^*$ was accomplished in a radionuclide neutron source ^{241}Am-Be, an ampoule containing a homogeneous mixture of americium (Am_2O_3) and beryllium. In this system the nuclei of the latter element are surrounded by americium nuclei and the alpha-particles emitted by the nuclei of ^{241}Am in any one direction bombard the beryllium nuclei, giving rise to the reaction $^9Be(\alpha, n)^{12}C^*$ and a subsequent emission of gamma-radiation with an energy of 4438 keV. The inelastic scattering of neutrons emitted from the Am-Be source, on carbon nuclei was studied in a geometrical arrangement depicted schematically in Fig. 2. The neutron source was surrounded by a carbon-containing material (graphite, brown coal, limestone, polyethylene) so that the neutrons emitted in all directions may bring about an inelastic scattering on carbon nuclei.

The form of the function $w(E'_\gamma)$ derived for such geometrical arrangements describes the energy distribution of the observed gamma-photons giving rise to the peak of the spectral line. Let the quantity $w(E'_\gamma)dE'_\gamma$ correspond to the probabi-

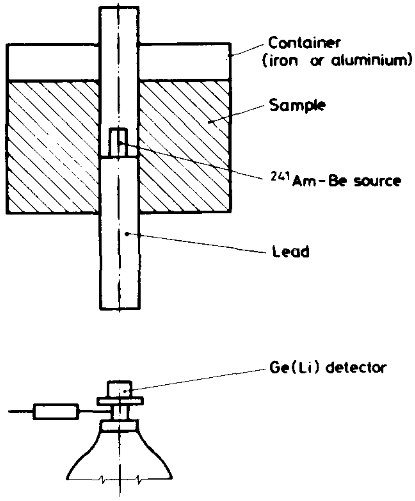

Fig. 2. Scheme of the experimental arrangement used in the study of inelastic neutron scattering on carbon

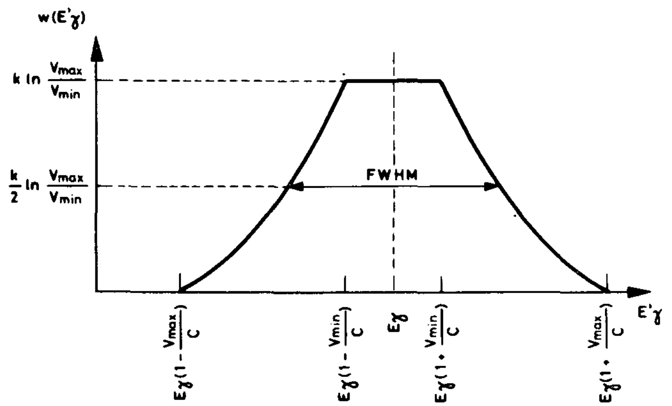

Fig. 3. The shape of the w(E'_γ) function

lity that the set of excited nuclei will emit a photon towards the detector with an energy E'_γ falling within the interval E'_γ, $E'_\gamma + dE'_\gamma$. Then function w(E'_γ) reflects the density of probability of the energy distribution of the photons in the peak. The function w(E'_γ) was derived under the following simplifying assumptions:

a) flight directions of the nuclei emitting gamma-rays have an isotropic spatial distribution,

b) the magnitudes of velocities of emitting nuclei are uniformly distributed in the interval $\langle v_{min}, v_{max} \rangle$,

c) the probability of gamma-photon emission by the nucleus is the same for all angles Φ,

d) the detector (observer) of the gamma-radiation is sufficiently far from the set of decaying nuclei.

Under these assumptions the function $w(E'_\gamma)$ has the form (Fig. 3):

$$w(E'_\gamma) = \begin{cases} 0 & , \quad E'_\gamma \leqslant E_\gamma \left(1 - \frac{v_{max}}{c}\right) \\ k \ln \frac{v_{max}}{\left(1 - \frac{E'_\gamma}{E_\gamma}\right) c} & , \quad E_\gamma \left(1 - \frac{v_{max}}{c}\right) \leqslant E'_\gamma < E_\gamma \left(1 - \frac{v_{min}}{c}\right) \\ k \ln \frac{v_{max}}{v_{min}} & , \quad E_\gamma \left(1 - \frac{v_{min}}{c}\right) \leqslant E'_\gamma \leqslant E_\gamma \left(1 + \frac{v_{min}}{c}\right) \\ k \ln \frac{v_{max}}{\left(\frac{E'_\gamma}{E_\gamma} - 1\right) c} & , \quad E_\gamma \left(1 + \frac{v_{min}}{c}\right) < E'_\gamma \leqslant E_\gamma \left(1 + \frac{v_{max}}{c}\right) \\ 0 & , \quad E_\gamma \left(1 + \frac{v_{max}}{c}\right) \leqslant E'_\gamma \end{cases} \tag{4}$$

where

$$k = \frac{c}{2E_\gamma} \cdot \frac{1}{v_{max} - v_{min}}.$$

Application of the function $w(E'_\gamma)$ makes it possible to derive parameters characterizing the shape of the spectral line peak. The mean value of energy of the observed photons is:

$$\overline{E'_\gamma} = \int_{E_\gamma(1 - \frac{v_{max}}{c})}^{E_\gamma(1 + \frac{v_{max}}{c})} E'_\gamma w(E'_\gamma) dE'_\gamma = E_\gamma \quad ,$$

variance is

$$D(E'_\gamma) = \sigma^2 = \int_{E_\gamma(1-\frac{v_{max}}{c})}^{E_\gamma(1+\frac{v_{max}}{c})} (E'_\gamma - \overline{E'_\gamma})^2 w(E'_\gamma) dE'_\gamma = \frac{1}{g} \left(\frac{E_\gamma}{c}\right)^2 (v^2_{max} + v_{max}v_{min} + v^2_{min}), \tag{5}$$

the peak width at a half maximal height is

$$FWHM = 2E_\gamma \sqrt{\frac{v_{max}}{c} \cdot \frac{v_{min}}{c}}\ , \tag{6}$$

the peak width at one tenth of the maximal height is

$$FWTM = 2E_\gamma \left[\left(\frac{v_{max}}{c}\right)^g \frac{v_{min}}{c}\right]^{\frac{1}{10}}, \tag{7}$$

and the total peak width is

$$\Delta E'_\gamma = 2E_\gamma \frac{v_{max}}{c}. \tag{8}$$

The minimum (v_{min}) and maximum (v_{max}) values of velocities of the excited nuclei of $^{12}C^*$ were calculated from the kinematics of the appropriate process.[1] These limit velocity values determine the interval encompassing the initial velocities of excited carbon nuclei. The photon emission takes place not only at the moment of formation of $^{12}C^*$ nuclei but also later on after a certain time interval. At this time the nucleus has a different, lower velocity than at the moment of formation since its movement through the medium entails ionization losses and attendant losses in kinetic energy. In a time interval of 0–3 τ (τ – mean life-time of the excited nucleus) about 95%of the excited nuclei emit a photon. For this reason, the velocities v'_{min}, v'_{max} of carbon nuclei after a flight time equal to 3 τ were determined by means of the ionization losses of carbon ions in the material.[2] The velocities v'_{min}, v'_{max}, v_{min}, v_{max} determined in this way yielded the intervals of maximum $< v'_{max}, v_{max} >$ and minimum $< v'_{min}, v_{min} >$ limit velocities at which 95% of the excited nuclei emit a gamma-photon. The widths of limit velocity intervals depend on the energy of particles giving rise to the process. In the case of the reaction $^9Be(\alpha, n)^{12}C^*$ the energy of the alpha-particles was

Table 1
Velocities of excited nuclei of carbon $^{12}C^*$

Reaction, process	Energy of incident particles, MeV	$\frac{v_{max}}{c}$	$\frac{v'_{max}}{c}$	$\frac{v_{min}}{c}$	$\frac{v'_{min}}{c}$	$\frac{\bar{v}_{max}}{c}$	$\frac{\bar{v}_{min}}{c}$
$^9Be(\alpha, n)\,^{12}C^*$	5.48	0.025	0.022	0.0083	0.0060	0.0235	
	3.0	0.019	0.016	0.0056	0.0045		0.00505
$^{12}C(n, n')^{12}C^*$	7.0	0.0147	0.0120	0.0042	0.0030	0.0134	0.0036
	6.0	0.0126	0.0100	0.0048	0.0038		
	5.0	0.0095	0.0080	0.0064	0.0050		

Velocities v_{max}, v_{min} were calculated according to Eq. (21) from the book of DECONNINCK,[1] p. 120. The theoretical values of parameters FWHM, FWTM and $\Delta E'_\gamma$ were calculated from velocities $\bar{v}_{max}$, $\bar{v}_{min}$ given in the last two columns. The mean lifetime of the excited carbon nuclei $\tau = 2.6 \cdot 10^{-14}$ s.[5]

Table 2
Calculated and experimental values of the overall width of gamma-radiation peak $\Delta E'_\gamma$ and parameters FWTM and FWHM for the reaction $^9Be(\alpha, n\gamma)^{12}C$ and the process $^{12}C(n, n'\gamma)^{12}C$

	$^9Be(\alpha, n)^{12}C$		$^{12}C(n, n'\gamma)^{12}C$	
	Theory	Experiment	Theory	Experiment
$\Delta E'_\gamma$, keV	209 ± 14	190 ± 7	119 ± 12	110 ± 8
FWTM, keV	179 ± 12	160 ± 4	104 ± 11	97 ± 6
FWHM, keV	97 ± 9	90 ± 4	62 ± 9	64 ± 8

Experimental values are given with the mean square errors.

chosen in the interval from 3.0 MeV (approximately the height of the potential barrier) to 5.48 MeV. In the case of $^{12}C(n, n'\gamma)^{12}C$ reaction a neutron energy interval was chosen from 5 to 7 MeV. The magnitudes of velocities for the two processes under study, with respect to the energy interval of the incident particles, are given in Table 1. The pairs of quantities v'_{min}, v_{min} and v'_{max}, v_{max} were used to calculate the mean velocities $\bar{v}_{min}$, $\bar{v}_{max}$ which were then inserted into Eqs (6), (7) and (8) to obtain theoretical values of the parameters FWHM, FWTM and $\Delta E'_\gamma$ which are shown in Table 2 for both the processes in question. The errors reflect the width of the limit velocity interval.

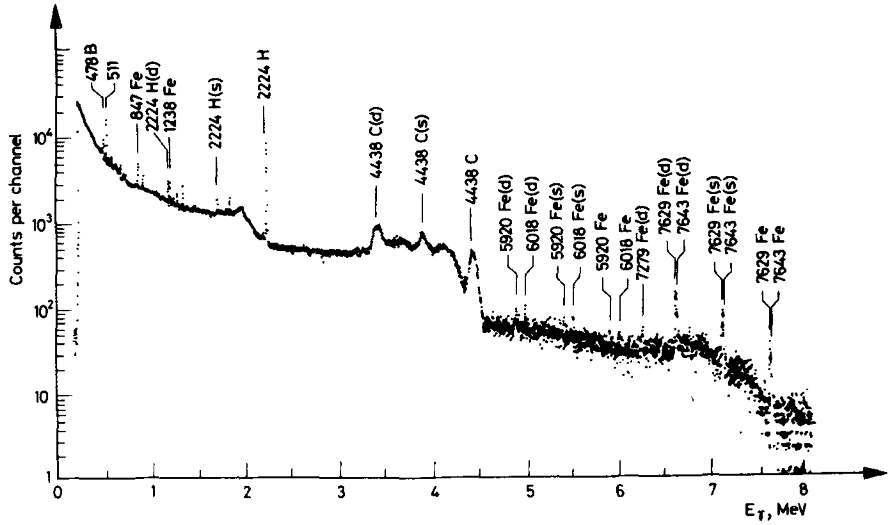

Fig. 4. Gamma-radiation spectrum excited by neutrons from a ^{241}Am-Be source and in the reaction ^{9}Be(α, nγ)^{12}C

Comparison of theoretical and experimental values

Fig. 4 shows the spectrum of gamma-radiation from Am-Be source, observed with a semiconductor Ge(Li) detector (energy resolution 7 keV for 7.5 MeV). The radionuclide Am-Be source was placed in a portable polyethylene container about one meter from the detector. The container was surrounded by a "Neutrostop"[6] shielding interspersed with iron blocks. The Ge(Li) detector was not shielded from the direct gamma-radiation emitted from the neutron source. The spectrum exhibited peaks corresponding to the gamma-radiation from a radiative capture of thermal neutrons by hydrogen (2224 keV) and iron (5920 keV, 6018 keV, 7629 keV, 7643 keV) nuclei, a peak of an annihilation radiation (511 keV) and three wide, Doppler-broadened peaks [4438C(d), 4438C(s), 4438C(f)] corresponding to the energy of the 4438 keV carbon line excited in the reaction ^{9}Be(α, n)^{12}C*; ^{12}C* $\longrightarrow$ ^{12}C + γ in the neutron source. The peak 4438C(f) corresponds to the full absorption of a 4438 keV photon in the Ge(Li) detector, peak 4438C(s) [and 4438C(d)] corresponds to the escape of one (or both) annihilation photons from the detector.[3] The detailed shape of the trio of carbon peaks measured with a high statistics (measurement time 50 000 s) is shown in Fig. 5. Full lines represent the theoretical shape of the peak of the spectral line calculated according to Eq. (4). From the measured peaks we determined the mean velocities $\bar{v}_{max}^{exp} = (2.14 \pm 0.08) \cdot$

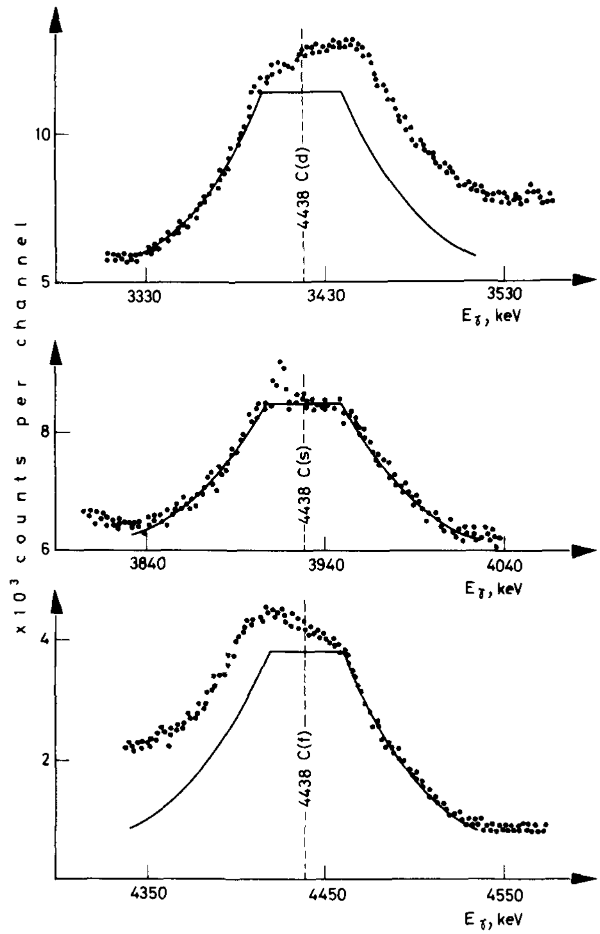

Fig. 5. Shapes of carbon peaks 4438C(f), 4438C(s), and 4438C(d)

$\cdot 10^{-2}$c and $\bar{v}_{min}^{exp} = (4.88 \pm 0.39) \cdot 10^{-3}$c, which are in a good agreement with the theoretical values of $\bar{v}_{max}$ and $\bar{v}_{min}$. The designated mean positions of the peaks 4438C(d), 4438C(s), and 4438C(f) were determined from the energy calibration of the spectrum. Peak 4438C(s) most closely resembles the theoretical distribution in its shape (Fig. 3). In the left part of the peak there arises the small interference peak 4934Si(d) from the capture of thermal neutrons by silicon contained in the walls of the measuring room. The asymmetry of the peak 4438C(d) is due to the contribution of the Compton scattering of the annihilation radiation into the right-hand side of the peak while the asymmetry of the peak 4438C(f) is due to

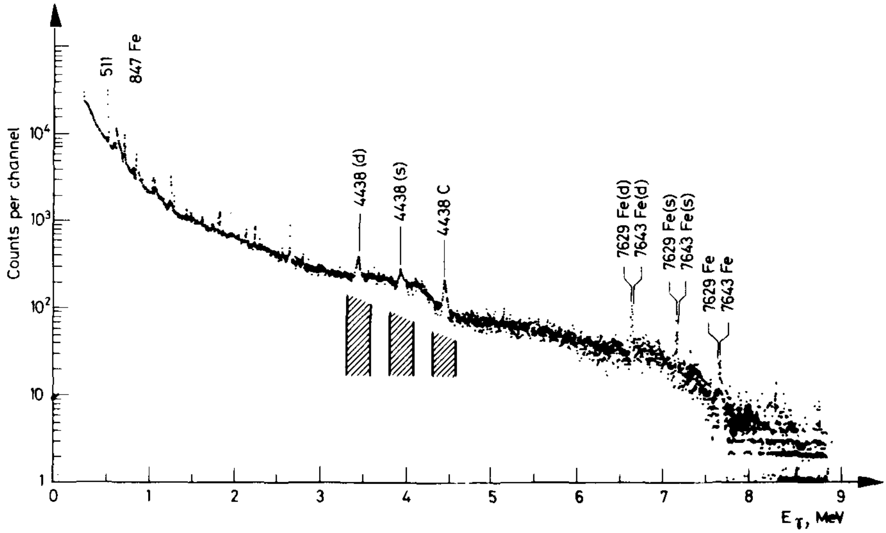

Fig. 6. Gamma-radiation spectrum excited in the inelastic neutron scattering on carbon

the contribution of multiple processes into its left-hand side.[4] The intact left-hand part of peak 4438C(d) and the intact right-hand part of peak 4438C(f), as well as the peak 4438C(s), were used to determine the values of FWHM, FWTM and $\Delta E'_\gamma$. The arithmetic mean of these values yielded the experimental values of FWHM, FWTM and $\Delta E'_\gamma$ shown in Table 2 together with calculated values. A comparison shows a good agreement of measured and calculated values.

The inelastic scattering of neutrons on carbon was studied in an arrangement shown in Fig. 2. The Ge(Li) detector was sufficiently shielded from the direct gamma-radiation due to the $^9Be(\alpha, n\gamma)^{12}C$ reaction in the Am-Be source by a 25 cm thick lead cylinder. One of the gamma-radiation spectra of graphite is shown in Fig. 6. The process also exhibits a Doppler broadening of peaks of the 4438 keV line, though somewhat less conspicuous than in the reaction $^9Be(\alpha, n\gamma)^{12}C$. The numerical values of the broadening, obtained as a mean of several measurements, are shown in Table 2 together with the calculated values; a comparison again showed a good agreement between the two sets of values.

It should be noted that the Doppler broadening of peaks of the 4438 keV line in the reaction $^9Be(\alpha, n\gamma)^{12}C$ is about 20-fold, in the case of the $^{12}C(n, n'\gamma)^{12}C$ about 15-fold bigger than the energy resolution of the Ge(Li) detector and is approximately equal to the energy resolution of scintillation detectors with the NaI(Tl) crystal. The natural width of the 4438 keV line is a mere 0.015 eV.

Conclusion

The Doppler broadening values obtained experimentally for the two processes agree well with the values calculated on the basis of kinematic considerations. The Doppler broadening due to numerous types of inelastic interactions with light nuclei may be observed by means of the Ge(Li) detectors whilst scintillation detectors cannot be used owing to the low energy resolution. Considered from the viewpoint of the analytical applications of the prompt gamma-radiation from inelastic interactions, the Doppler broadening, if bigger than the resolution of the detector, brings about a lowering of the analytical sensitivity. On the one hand it enhances the possibility of energy interferences while on the other, it may serve as a complementary information in the determination of the origin of the line.

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