IOP Publishing Physica Scripta
Phys. Scr. **79** (2009) 065007 (6pp) doi:10.1088/0031-8949/79/06/065007

A Mössbauer experiment in a rotating system on the second-order Doppler shift: confirmation of the corrected result by Kündig

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Received 16 January 2009
Accepted for publication 6 April 2009
Published 27 May 2009
Online at stacks.iop.org/PhysScr/79/065007

Abstract
We present the results of a Mössbauer experiment in a rotating system, whose performance was stimulated by our recent findings (2008 *Phys. Scr.* **77** 035302) and which consisted of the fact that a correct processing of Kündig's experimental data on the subject gives an appreciable deviation of a relative energy shift $\Delta E/E$ between emission and absorption resonant lines from the standard prediction based on the relativistic dilation of time (that is, $\Delta E/E = -v^2/2c^2$ to the accuracy c^{-2}, where v is the tangential velocity of the absorber of resonant radiation, and c is the velocity of light in vacuum). That is, the Kündig result we have corrected becomes $\Delta E/E = -k(v^2/c^2)$, with $k = 0.596 \pm 0.006$ (instead of the result $k = 0.5003 \pm 0.006$, originally reported by Kündig). In our own experiment, we carried out measurements for two absorbers with a substantially different isomer shift, which allowed us to make a correction of the Mössbauer data regarding vibrations in the rotor system at various rotational frequencies. As a result, we obtained the overall estimation $k = 0.68 \pm 0.03$.

(Some figures in this article are in colour only in the electronic version.)

1. Introduction

In our recent paper [1], we found that a familiar experiment by Kündig on the transverse Doppler shift in a rotating system measured with the Mössbauer effect [2] contained errors in the data processing. An intriguing fact is that after correction of the errors by Kündig, his experimental data give the value [1]

$$\frac{\Delta E}{E} = -k\frac{v^2}{c^2}, \qquad (1)$$

where $k = 0.596 \pm 0.006$, instead of the standard relativistic prediction $k = 0.5$. One can see that the deviation of the coefficient k in equation (1) from 0.5 exceeds by almost 20 times the measuring error. It is important that the revealed deviation cannot be attributed to the influence of rotor vibrations and other disturbing factors, since all these factors have been excluded by a perfect methodological trick applied by Kündig: a first-order Doppler modulation of the energy of γ-quanta on a rotor at each fixed rotation frequency. Due to this Kündig's experiment should be classified as the most reliable among other experiments on the subject [3–7], where the experimenters measured only the count rate of detected γ-quanta as a function of rotation frequency. Furthermore, we have shown in [1] that the experiment [7], containing much more data than [3–6], is also well fitted into the supposition $k > 0.5$.

These findings stimulated us to carry out our own experiment on the time dilation effect in a rotating system (or the second-order Doppler shift, (SOD)), which is described below in section 2.

Planning a scheme of this experiment, we recognized that a direct repetition of the Kündig experiment would leave some doubt on the presence of some missed technical factors, which distort the measured value of k. One such factor might be the finite length (about 1 cm) of the piezotransducer applied in the Kündig experiment. Hence, some parts of the piezotransducer inevitably experience the centrifugal force during a rotation, when its face lies on the rotation axis. One cannot exclude that this could change the piezoelectric constant with variation of the rotation frequency. Although Kündig estimated this factor to be negligible [2], he did not present a convincing proof. Therefore, we decided to repeat neither the scheme of the Kündig experiment nor the schemes of other known experiments on the subject mentioned above [3–7], in order to get independent information on the value of k in equation (1). In particular, we refrained from the first-order Doppler modulation of the energy of γ-quanta, in order to exclude the uncertainties in the realization of this method, as mentioned above. Thus we followed the standard scheme [3–7], where the count rate of detected γ-quanta N as a function of the rotation frequency ν is measured (section 2.1). However, in contrast to the experiments [3–7], we do evaluate the influence of chaotic vibrations on the measured value of k. For this purpose, we developed a method, which involves a joint processing of the data collected for two selected resonant absorbers with the specified difference of resonant line positions in the Mössbauer spectra (section 2.2). The results we obtained confirmed the original Kündig result, following the correction we brought to it, that the coefficient k in equation (1) substantially exceeds 0.5 (section 2.3).

2. Mössbauer experiment in a rotating system with two resonant absorbers

2.1. Experimental setup

The rotor system developed by us is based on the ultracentrifuge K-80 (Belmashpribor, Minsk) with the diameter of the working chamber 630 mm and the range of rotation frequencies $\nu = 0$–150 rev. s^{-1}. The rotor has the form of a flat streamlined rod, and it was made from a special ultrastrong and light aluminum alloy doped by titanium and exposed to special thermal treatment. The diameter of the rod is 610 mm. A standard Mössbauer source ^{57}Co(Cr) (Ritverc, S.-Petersburg, Russia) with the activity of 20 mCi was put into a Cu–Pb shielding and collimating system and mounted on the rotation axis. A sample holder made of a special tempered aluminum alloy was fixed at the edge of the rotor. A semi-hermetic chamber of the rotor system was continuously pumped out during measurements; the stationary pressure was about 100 mmHg. In these conditions there was some heating of the rotor during its rotation. However, the difference between the temperatures of the source and absorber never exceeded 10 °C within the applied range of rotation frequencies. For such a difference of temperature, we can neglect the variation of the resonant line position due to the thermal shift.

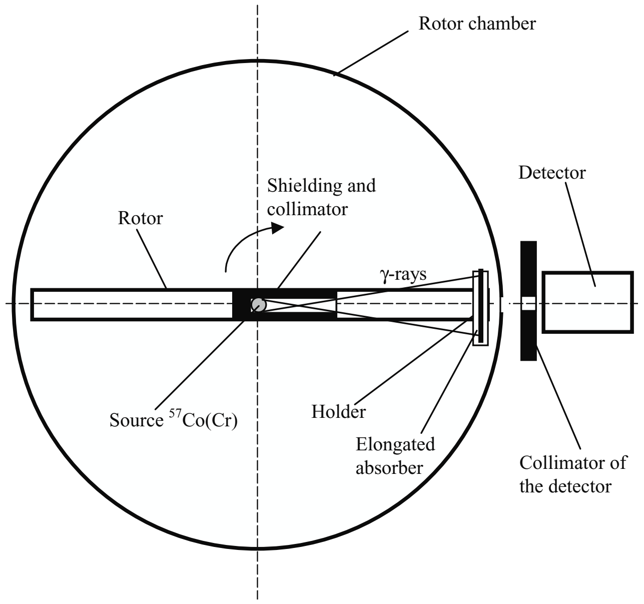

Figure 1. Schematic of our experiment.

A proportional counter for the detection of γ-quanta, filled with xenon, was located outside the rotor system. A beryllium window (Ø = 15 mm) for the output of resonant radiation was made in the working chamber opposite, the working window of the detector, and its center belonged to the same plane as the line joining the source and the absorber. For the reasons explained below (see section 3), we chose the distance between the source and the absorber ($r = 305$ mm) many times larger than that chosen for the Kündig experiment (where $r = 46$ mm). Then the maximum tangential velocity at the edge of the rotor is $v = 2\pi\nu r = 2\pi \times 150 \times 0.305 = 287.3$ m s^{-1}.

In order to partially compensate for the decrease of count rate of the detector, caused by the enlarged distance between the source and the absorber, we applied the absorbers elongated in the azimuthal direction. It allows us to increase the effective measuring time per rotation period as much as possible (see figure 1). The absorbers used had a rectangular shape the size of 15×55 mm^2. In these conditions the average count rate of the detector was about 3 pulses s^{-1} in the working energy window, selecting resonant γ-quanta of ^{57}Fe 14.4 keV. Due to a careful shielding of the source, the background count rate (measured at the angular positions of the rotor, when radiation of the source does not pass through the detector's window) was less than 0.2 pulses s^{-1}. This allowed us to omit the time selection of output pulses of the detector, applied in the earlier experiments [2–7].

A measurement of SODs was carried out in the range of rotation frequencies $\nu = 70$–120 rev. s^{-1}. This corresponds to the change of tangential velocities of the absorber from $v = 134.1$–230.0 m s^{-1}. In terms of the first-order Doppler shift, it corresponds to the change of linear velocity

$$u = c\frac{v^2}{2c^2} = (0.030\text{–}0.088)\ \text{mm s}^{-1}\ (\text{for } k = 0.5).$$

Each measuring cycle started with the maximal frequency $\nu = 120$ rev. s^{-1}, with its further decrease to 70 rev. s^{-1} in steps of 10 rev. s^{-1}. The accuracy of setting ν is ±0.5 rev. s^{-1}.

The number of output pulses of the detector was measured during 100 s at each rotational frequency. Then a new measurement cycle was started, and so on. The total number of counts at each ν has been obtained by summing over 50 cycles.

2.2. Methodology

In what follows, we consider k in equation (1) as a parameter to be determined experimentally. In an ideal case, where the mechanical vibrations are absent in the rotating system, the coefficient k in equation (1) is easily computed, if we compare the SOD, obtained as a function of the rotation frequency, with the first-order Doppler shift to be measured for the same pair of 'source plus absorber' with a standard Mössbauer spectrometer outside the rotor system.

Further on we proceed from the fact that the non-vanished vibrations are always present in the rotating system. Such vibrations are known to broaden the given resonant line, but do not influence the total area and position of this resonant line on the energy scale. In a general case, we have to assume that the vibrations are not only present in the system, but also that the vibration level depends on the rotation frequency. Just such a behavior of vibrations has been found by Kündig (a broadening of resonant line up to 1.5 times with an increase of rotation frequency for the full range of its variation from 11 000 to 35 000 rpm [2]). In this case the count rate of the detector N, as a function of the rotation frequency ν, can essentially deviate from the corresponding dependence $N(\nu)$ in the ideal case (no vibrations), and accordingly, one may end up with distorted information on the value of k in equation (1).

In order to eliminate such a distortion of k due to vibrations, we applied two different resonant absorbers with substantially different parameters of the Mössbauer spectra. Correspondingly, the broadening of resonant lines due to vibrations should induce essentially different variations of the detector's count rate for each absorber. Thus, comparing the data of the rotor experiment obtained with each absorber and supposing an equal level of vibrations for both absorbers (they have equal sizes and masses with the precision better than 0.1%), we can get full information on the level of vibrations and make the required corrections under evaluation of k.

In practice, it is enough to deal with two resonant lines: one of them has a maximum within the full range of variation of the energy (absorber 1, figure 2(a)), and the second line has an appropriately larger isomer shift than for the first absorber, so that the full range of variation of the energy of γ-quanta due to the SOD lies on a slope of the line (absorber 2, figure 2(b)). Now let us show that a conjoined processing of data obtained in a rotor experiment with absorbers 1 and 2 allows us to actually eliminate the influence of vibrations on the estimation of k. For this purpose we realize an algorithm as follows:

(1) The Mössbauer spectra of both absorbers are measured with a high statistic quality.
(2) The data of the rotor experiment are collected for absorbers 1 and 2 in the same range of variation of rotation frequency.
(3) Having assumed any particular value of k in equation (1) (for example, one can vary k between 0.5 and 1.0), the expected theoretical curves $N(\nu)$ for a rotor experiment with a zero level of vibrations (hereinafter the 'idealized rotor experiment') are plotted for both absorbers. Since in a real experiment the vibrations are expected to be present anyway, the theoretical curves we draw for each absorber deviate from corresponding experimental data, no matter what the assumption on k is.
(4) For the chosen value of k, a variation of the width Γ for the resonant line of absorber 2 is implemented at each rotation frequency ν, so that the corrected theoretical curve $N(\nu)$ passes exactly through the available experimental points. As an outcome of this procedure we obtain the dependence $\Gamma(\nu)$, which models the line broadening due to vibrations at various ν.
(5) The dependence $\Gamma(\nu)$ obtained at the given k is applied to correct the theoretical curve for absorber 1, obtained for the same k. If this new curve continues to deviate from a set of corresponding experimental points, then we assume that the hypothesis on a given value of k is false and should be rejected.
(6) A new value of k is chosen, and steps 3–5 are performed again, while we obtain a self-consistent result for absorber 1 with the minimal statistical test criterion χ^2. A corresponding magnitude of k is then adopted.

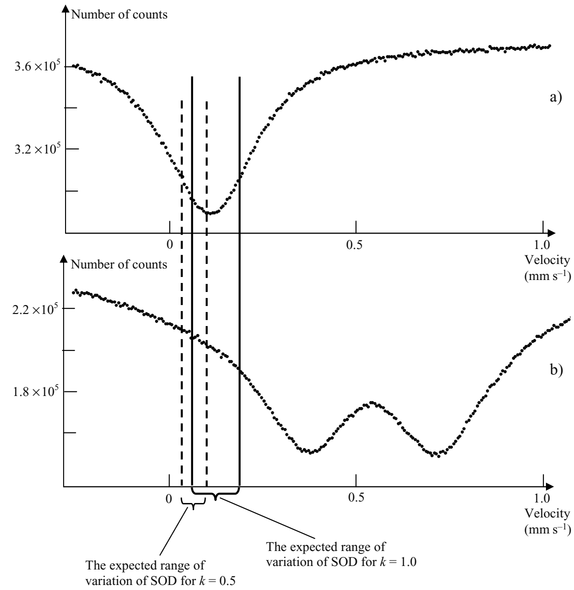

Figure 2. Mössbauer spectra of absorber 1 ($K_4\,{}^{57}Fe(CN)_6 \times 3H_2O$), (a) and absorber 2 ($Li_3\,{}^{57}Fe_2(PO_4)_3$), (b), obtained with the source $^{57}Co(Cr)$, and the expected range of variation of SODs in our rotor experiment for two limited hypotheses on k.

As a result we get the best fitting of experimental data with two sets of free parameters: the coefficient k in equation (1) and the level of vibrations, manifesting as $\Gamma(\nu)$ dependence. In section 2.3 we use this algorithm for processing the data obtained, applying the MathCad Professional software.

We would like to add that a modern extended database on various Mössbauer compounds created by the Mössbauer Effect Data Center [8] allows us to choose the most optimal absorbers for realization of the algorithm just described. Absorber 1 represents a thin layer of the compound $K_4Fe(CN)_6 \times 3H_2O$ enriched with ^{57}Fe to 90%. Absorber 2 is a thin layer of the compound $Li_3Fe_2(PO_4)_3$ enriched

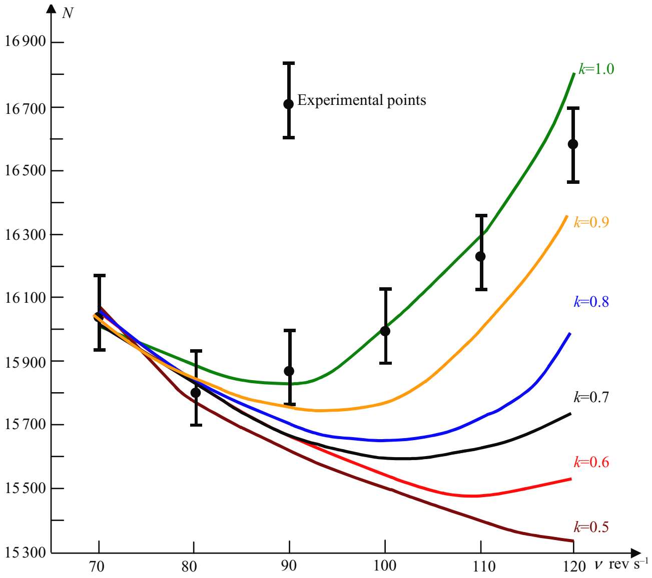

Figure 3. Experimental data for absorber 1 in comparison with the curves computed at different k for the idealized (no vibrations) rotor experiment.

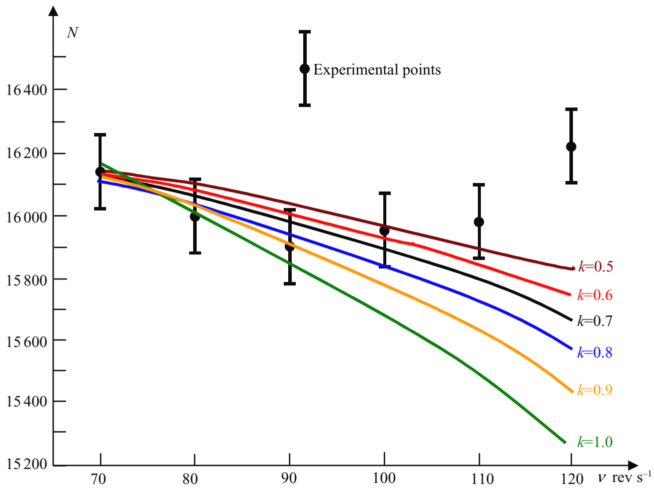

Figure 4. Experimental data for absorber 2 in comparison with the curves computed at different k for the idealized (no vibrations) rotor experiment.

with ^{57}Fe to 90%. Each absorber was placed into beryllium shielding transparent for resonant γ-quanta 14.4 keV. The Mössbauer spectra of absorbers were measured by means of the Mössbauer instrument package MS-2000IP [9].

2.3. Results

In figure 2, we have already shown the Mössbauer spectra of absorbers 1 (figure 2(a)) and 2 (figure 2(b)), where the dotted vertical lines restrict a range of variation of SODs for $\nu = 70$–120 rev. s^{-1} and $k = 0.5$, whereas the continuous vertical lines show the same range for $k = 1.0$. The spectrum of the first absorber represents a single line shifted at $+(0.095 \pm 0.001)$ mm s^{-1} with respect to the emission line of ^{57}Co(Cr). The value of the resonant effect is (20.9 ± 0.1)%. The spectrum of the second absorber represents a partially resolved doublet with the left line shifted at $+(0.390 \pm 0.001)$ mm s^{-1} with respect to ^{57}Co(Cr). The resonant effect is (30.5 ± 0.1)%.

One can see that the parameters of the Mössbauer spectrum of absorber 1 make it especially sensitive to the choice of two limited cases $k = 0.5$ and 1.0, if one considers the idealized rotor experiment (exempt from vibrations). If $k = 0.5$, then the count rate of the detector should continuously decrease in the full range of variation of the rotation frequency ν, and at $\nu = 120$ rev. s^{-1} we only approach to the minimum of resonant absorption. If $k = 1.0$, then the count rate of the detector reaches a minimal value already at $\nu \approx 90$ rev. s^{-1} with a further increase at higher ν.

A similar analysis of the idealized rotor experiment with absorber 2 indicates that the count rate of the detector decreases for both the limited cases ($k = 0.5$ and 1.0), but a slope of the falling curve is a few times larger at $k = 1.0$.

In figures 3 and 4 we present the data of our rotor experiment, obtained with both absorbers. In these figures we also plotted the corresponding curves, expected in the idealized rotor experiment at various k. According to our approach to the data processing described above (section 2.2), we use the data of figure 4 (absorber 2) to evaluate the level of vibrations in the rotor system at different υ. Then we recompute the expected curves for absorber 1 at various k with account of vibrations in the system and compare them with the experimental data of figure 3.

Analysis of figure 4 (absorber 2) indicates that vibrations are actually present in the system (like they were in the Kündig experiment) and do distort the count rate of the detector in comparison with the idealized case. In particular, the number of counts obtained at $\nu = 120$ rev. s^{-1} does not comply with the computed curves in the full adopted range of variation of $k = 0.5$–1.0. Furthermore, since these computed curves have a different slope for different k, the level of vibrations expressed through the dependence $\Gamma(\nu)$ is sensitive to the choice of a particular k. At the same time, at this stage we can reduce the volume of computing work by rejecting the two limited hypotheses $k = 0.5$ and $k = 1.0$ as physically non-adequate, assuming that just one k is to be adopted throughout. Indeed, if we suppose the validity of $k = 0.5$ (this is just the classical relativistic prediction) then we conclude from the data of figure 4 (the experiment with absorber 2) that up to $\nu = 110$ rev. s^{-1}, the vibrations in the rotor system are negligible. In such a case the experimental points depicted in figure 3 (the experiment with absorber 1) must lie on the curve computed for $k = 0.5$ at least in the range $\nu = 70$–110 rev. s^{-1}. However, we observe a drastic deviation of the experimental data from this curve. Furthermore, if we assume that the data of the experiment with absorber 1 (figure 3) match well with the computed curve for $k = 1.0$, we must conclude that the level of vibration is negligible in the full range of variation of ν. However, a comparison of the computed curve for absorber 2 at $k = 1.0$ with experimental data (figure 4) shows that vibrations in the rotor system should be relatively high for $\nu \geqslant 90$ rev. s^{-1}. Hence we again get a self-contradictory result. Thus, we restricted our further analysis to the range $k = 0.6$–0.9.

In figure 5 we again present the results of the rotor experiment with absorber 1 in comparison with the expected theoretical curves recomputed for $k = 0.6$, 0.7 and 0.8 by

Table 1. Relative broadening of the resonant line at various rotation frequencies ν. The line widths at $\nu = 100$, 110 and 120 rev. s^{-1} are directly computed from the difference between the experimental data obtained with absorber 2 and the expected theoretical curves $N(\nu)$ for an idealized rotor experiment (figure 4) at $k = 0.7$. The line widths at $\nu = 70$, 80 and 90 rev. s^{-1} have been obtained by extrapolating the revealed exponential law (to be found also in the Kündig experiment [2]) of line broadening as a function of ν in the range 100–120 rev. s^{-1}, to the range of $\nu = 70$–90 rev. s^{-1}. Such an extrapolation has been applied, since a deviation of the $N(\nu)$ curve from the experimental data at these lower frequencies is less than the statistical error of the data.

Rotational frequency (rev. s^{-1})	70	80	90	100	110	120
Line width (in relative units)	1.01 ± 0.01[a]	1.02 ± 0.01[a]	1.04 ± 0.01[a]	1.08 ± 0.01[b]	1.15 ± 0.02[b]	1.30 ± 0.03[b]

[a]Extrapolated.
[b]Computed from data.

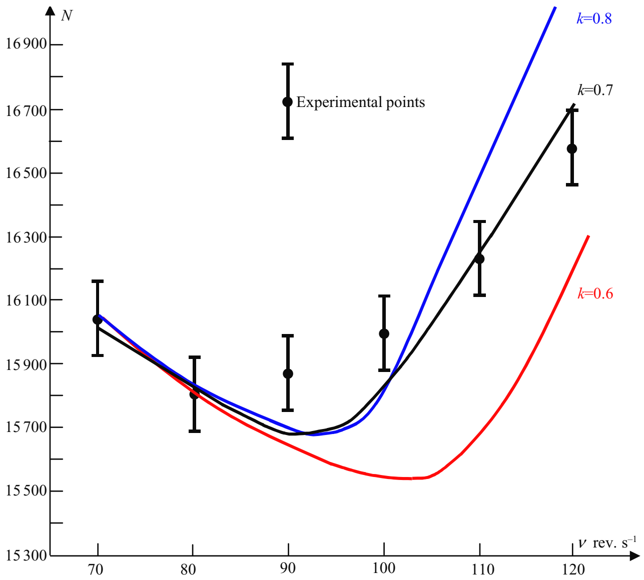

Figure 5. Experimental data for absorber 1 and the expected curves recomputed at different k taking into account the level of vibrations in the rotor system.

taking into account the dependence $\Gamma(\nu)$, obtained from the data processing with absorber 2. It can be clearly seen that the best correspondence between the experimental data and theoretical curve occurs at nearly $k = 0.7$. A least square fit specifies this observation to lead to $k = (0.68 \pm 0.03)$.

Finally, in table 1 we show a relative broadening of resonant line at various rotation frequencies ν and $k = 0.7$. The maximal broadening of resonant line is about 30% at $\nu = 120$ rev. s^{-1} (tangential velocity $v = 230\,\mathrm{m\,s^{-1}}$), which is comparable with the line broadening in the Kündig experiment at the same tangential velocity.

3. Conclusion

As in the corrected processing of the data of Kündig's experiment, we have the privilege of having made a contribution to [1]; once again we reveal an appreciable deviation of the coefficient k in equation (1) from the classical relativistic prediction $k = 0.5$. Of course, we trust in the validity of the usual relativistic dilation of time due to the motion, which has numerous confirmations in the experiments dealing with atomic beams and free muons (see, e.g. [10, 11]). Rather, we conjecture that with reference to the Mössbauer experiments on a rotor, the energy shift of the absorption resonant line is induced not only via the standard time dilation, but also via some additional effect missed at the moment. Discussing in [1] a possible origin of this effect, we supposed that the pressure created by the centrifugal force on the absorber might change the electron density on the resonant nucleus and, by such a way, change the isomer shift between emission and absorption lines. This leads to the increase of measured k. Although this effect was not supported by numerical estimations of [1], we decided to eliminate any speculations on the influence of pressure, choosing the rotor diameter 6–7 times larger than in Kündig's and other Mössbauer experiments in rotating systems. In this case the centrifugal pressure on the absorber is the same times smaller for a fixed tangential velocity v. Nonetheless, we observe even a higher value of k than drawn by the Kündig experiment. This provides us with reason to assume that the missed effect, inducing the increase of k in comparison with the standard value 0.5, might have deep physical roots. The discrepancy between Kündig's and our result in the measurement of k allows us to suppose that k is not a constant value for various realizations of Mössbauer experiments in a rotating system, but, perhaps, increases with the decrease of a centrifugal pressure at the given tangential velocity. Of course, a complete understanding of the behavior of k will be achieved only after the development of a suitable theory. At the present stage, the implementation of new Mössbauer experiments in rotating systems seems to be the essential task, in order to collect new data on the relative energy shift between emission and absorption lines in various conditions. Planning such new experiments, one should recognize that the approach of Kündig (the first-order Doppler modulation of resonant radiation on a rotor) remains the best and does not lead to any instrumental errors; one can also see that the measuring error of k in his experiment is a few times lower than that in the reported experiment. This is not only due to a better statistic quality of Kündig's data obtained for the higher count rate of the detector, but also due to the elimination of any correction of these data to a level of vibration in a rotor system. At the same time, we hope that the present experiment, among other things, has its own independent significance as a definite confirmation of the corrected result of Kündig's experiment $k > 0.5$. It is fair to stress that the whole work was induced by the controversial prediction made by Yarman *et al* (see [11] and [12] of [1]), according to which k is expected to be larger than 0.5.

Acknowledgments

We thank Okan University (President of the Board of Trustees Bekir Okan and Rector Professor Sadik Kirbas; Istanbul, Turkey) and Savronik (Defense Electronics) Company (President Mustafa Kula and Vice President

Kenan Isik, old students of Professor T Yarman; Eskisehir, Turkey) for the financial support they have kindly provided (contract No. 08927 with the Belarus State University). We thank Professor Metin Arik for many hours of valuable discussions. Thanks are further due to Dr S Yarman and Dr F Yarman, Ex-Presidents of Savronik, for having honorably backed up the endeavor in question. We thank Eng. J Sobolev, Eng. V Furs (Belmashpribor, Minsk, Belarus) and Eng. A Zhenevskii (retired, Minsk, Belarus) for installation of the rotor system. We thank Dr M Silin and V Soloviov (V.G. Khlopin Radium Institute, S.-Petersburg, Russia) for preparation of both the absorbers. We thank Dr A Khasanov and Professor J Stevens (Mössbauer Effect Data Center, Asheville, USA) for extensive consultations regarding the choice of resonant absorbers for our research. Finally, we thank an anonymous referee for useful remarks, which were very helpful in the improvement of this paper.

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