Induced acceleration of the decay of the 31-yr isomer of $^{178m2Hf}$ using bremsstrahlung radiation

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A R T I C L E   I N F O

Article history:
Received 28 April 2015
Received in revised form 27 July 2015
Accepted 23 August 2015
Available online 28 August 2015

Editor: V. Metag

Keywords:
Nuclear isomers
$^{178m2Hf}$
Induced triggering
Electromagnetic transitions
Bremsstrahlung radiation

A B S T R A C T

$^{178m2Hf}$ isomer triggering was studied using the new experimental setup developed at the Kharkiv National University and installed at the Kyiv Institute for Nuclear Research. The target presenting a single Ta foil of 300 $\mu$m thickness with the $^{178m2Hf}$ isomer activity of 100 Bq was irradiated by 30 keV electron beam. The enhanced counting rates of all strongest ground-state band (with the energies of 213, 325 and 426 keV) and 8$^-$-state band (with the energies of 216, 495 and 574 keV) transitions from the $^{178m2Hf}$ isomer decay were observed. Our data are consistent with the total triggering effect of 1.55 $\pm$ 0.12%. An estimate for the photon induced triggering cross-section gives a value which is close to the upper limit obtained in the earlier published works on the $^{178m2Hf}$ isomer triggering.

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1. Introduction

Nuclear isomers present the unique form of energy storage. Decaying electromagnetically by $\gamma$-ray emission and electron conversion the nuclear isomers provide additional opportunities for stimulating the decay of isomers. Such release on demand or triggering of energy stored in nuclear isomers might lead to a broad range of applications including the controlled $\gamma$-ray sources and $\gamma$-ray lasers [1].

The $^{180m}Ta$ isomer is the only natural nuclear isomer. The triggering of the $^{180m}Ta$ isomer by 2.8 MeV [2] and less energetic [3] photons is reliably established. The physical interpretation for the triggering process is given in [4]. However, while the excitation energy of the $^{180m}Ta$ isomer is only 75 keV, the practical value of such triggering is minimal and presents only the fundamental interest. Much more effective and lower energy method how to release its energy should be discovered.

From 1998 all the efforts to accelerate the nuclear isomers decay were focused on the $^{16}^+$ 4-quasiparticle (QP) state of the nucleus $^{178}Hf$. A K isomer with the excitation energy $E_x = 2.4474$ MeV and half-life $T_{1/2} = 31$ years. A series of experiments were performed on this hafnium isomer with contradictory results [5,6, and references therein]. The most recent $^{178m2Hf}$ isomer triggering experiments were claimed to be either successful [7,8] or unsuccessful [9]. The theoretical papers either demonstrate no perspective for $^{178m2Hf}$ isomer depletion by low-energy X-rays [10, and references therein] or prove the possibility of such process [11, 12] to be observed experimentally. Anyway, the real mechanism of the triggering and its detailed scheme are not quite clear yet.

There are at least several well-known highly excited (about 1 MeV and higher) and long-lived ($T_{1/2} > 25$ d) nuclear isomers, among which the $^{178m2Hf}$ isomer is absolutely unique isomer, since it has both the highest excitation energy and the longest half-life. The uniqueness of the $^{178m2Hf}$ isomer has several very essential disadvantages. First, the production of this isomer is an extremely difficult task [13–15]. Second, the more productive nuclear reaction is chosen, the longer (in most cases from 6 to 20 years) cooling time for the by-products activity reduction is required. Such situation forced the intense search for any Ta targets irradiated by high-energy projectiles many years ago [16]. As a result, Ta foil of 300 $\mu$m thickness with about 100 Bq $^{178m2Hf}$ isomer activity that in 1967–1970 had been exploited as the converter at the Kharkiv 1.2 GeV Linac was found [17]. A set of 100 $\mu$m Ta foils with the $^{178m2Hf}$ isomer activity irradiated in 1987 by 100 MeV $\alpha$-particles was recently found at the Kyiv Institute for Nuclear Research [7].
Fig. 1. The scheme of spontaneous and induced decay of the $^{178m2}$Hf isomer.

The objective of this work is to check much more carefully which transitions observed in the spontaneous decay of the $^{178m2}$Hf isomer can really be enhanced.

2. Experimental details and results

The given $^{178m2}$Hf isomer triggering experiment was performed using the strongest of two available isomeric targets. While the $\gamma$-ray spectrum of the Kharkiv isomeric target is much more complicated compared to the weak (though practically not contaminated) Kyiv isomeric target, it allows acquiring much better statistics in the strongest $\gamma$-ray peaks of the $^{178m2}$Hf isomer for rather short life time of our cathodes (usually less than 4 hours at the maximum currents).

The experimental setup was developed at the Kharkiv National University and installed at the Kyiv Institute for Nuclear Research. Initially, the setup allowed irradiating the isomeric targets directly by electrons (or by X-rays when corresponding converters are used) with the energy 1–25 keV and current 0–150 μA. A little later the experimental setup was upgraded to the energy 1–30 keV and current 0–250 μA.

In this experiment the additional collimator present in our previous experiments [7,8] was removed allowing to obtain the electron currents up to 940 μA. At the same time, the diameter of the beam spot at the target enlarged from 8 mm (around the same size as the areas of two available spots each with practically the same $^{178m2}$Hf isomer activity) to 13 mm covering completely all the $^{178m2}$Hf isomer activity in the target.

The $\gamma$-ray spectra were acquired by HPGe coaxial detector mounted on the opposite side of the target in the horizontal plane and at 180° to the horizontally incident electron beam. In our experiment the HPGe detector GC 1818/5 (CANBERRA) and the standard acquisition system based on InSpector 2000 (CANBERRA) unit were used. The detector efficiency is 19.6% and its energy resolution at the $\gamma$1332 keV peak of $^{56}$Co is 1.67 keV. While the $\gamma$-ray self-absorption in the target is inevitable factor, the absorption in the experimental setup cover was ensured to be minimal and for that the Al cap of 2 mm thickness was used. The distance from the detector front face to the setup cap was about 2 mm. The decay scheme of the $^{178m2}$Hf isomer is shown in Fig. 1. The usual path of decay is to cascade down the 8− band until the 8−-state at the bandhead having 2-QP structure and a 4 s half-life is reached. The acquisition rate in the $\gamma$213 keV peak of the $^{178m2}$Hf isomer was around 4 counts per second. One of the $\gamma$-ray spectra with all strong peaks of the $^{178m2}$Hf isomer and other available sources in the target (131Ba and 150Eu) acquired in this experiment is shown in the Fig. 2.

All $\gamma$-ray spectra were processed by the WinSpectrum code [19]. As a result, accurate values for $\gamma$-ray peak areas and energies were obtained. Additionally, all $\gamma$-ray peaks were analyzed very carefully for consistency with the tabulated quantum yields [20]. Thus, any possible overlapping of recorded $\gamma$-ray lines was kept under complete control.

The entire series of measurements involved 20 runs of beam-on measurements at maximally possible (a little different for each cathode) electron currents. The acquisition period of such beam-on measurements was from around 2000 to 8000 seconds. The total irradiation time reached about 70000 seconds or 19 hours. In order to monitor the stability of measurement geometry, several runs of beam-off measurements were conducted before and after every irradiation. Such beam-off measurements accumulated data over the periods from a few hours to several days.

The beam-induced decay of the isomer resulted in the increase of intensities for all strongest ground-state band (with the energies of 213, 325 and 426 keV) and 8−-state band (with the energies of 216, 495 and 574 keV) transitions from the $^{178m2}$Hf isomer decay compared to the beam-off measurements (see Table 1).

Table 1

<table>
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<tr>
<th>Transition</th>
<th>Enhancement (%)</th>
<th>Uncertainty (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\gamma$213 keV (GBS)</td>
<td>2.033</td>
<td>0.268</td>
</tr>
<tr>
<td>$\gamma$216 keV (8− band)</td>
<td>1.444</td>
<td>0.294</td>
</tr>
<tr>
<td>$\gamma$325 keV (GBS)</td>
<td>1.469</td>
<td>0.259</td>
</tr>
<tr>
<td>$\gamma$426 keV (GB)</td>
<td>1.269</td>
<td>0.305</td>
</tr>
<tr>
<td>$\gamma$495 keV (8− band)</td>
<td>0.815</td>
<td>0.353</td>
</tr>
<tr>
<td>$\gamma$574 keV (8− band)</td>
<td>2.116</td>
<td>0.346</td>
</tr>
<tr>
<td>Averaged value</td>
<td>1.545</td>
<td>0.121</td>
</tr>
</tbody>
</table>

Fig. 2. $\gamma$-ray spectrum of our $^{178m2}$Hf isomeric target acquired during 4000 s. The energy ranges for planes (a) and (b) are chosen as it was done in [18] to demonstrate that the $^{178m2}$Hf isomer $\gamma$-ray peaks are not influenced by any other $\gamma$-ray peaks.
While there are no clear evidence that the induced decay of the $^{178m2}$Hf isomer populates different excited levels of $^{178}$Hf (see Fig. 3), the total counting rate of above-mentioned cascade transitions was used as a measure of the enhancement factor. The enhancement factor was estimated as 1.55 ± 0.12%. Thus, the significance of the triggering effect obtained in the present experiment is almost 13 standard deviations.

In our earlier paper [8], before the additional collimator was removed, due to much lower electron currents (up to 250 µA instead of up to 940 µA now) and smaller diameter of the beam spot at the target not covering completely all the $^{178m2}$Hf isomer activity in the target, the enhanced counting rates of only ground-state band (GSB) transitions were observed. The total counting rate of $8^+$-state band transitions was used to estimate the non-enhancement factor for these reference transitions. Since the statistics acquired in the $8^+$-state band transitions of $^{178m2}$Hf isomer turns out to be less than in the GSB transitions, the strongest transitions of all available sources in the target ($^{172}$Lu, $^{133}$Ba, and $^{150}$Eu) could also be taken into account. In this case the null enhancement factor was reliably obtained guaranteeing that the measurement geometry is stable during all the irradiations. Now when in the given experiment the enhanced counting rates of all strong GSB and $8^+$-state band transitions were observed, we can use as the reference transitions only the strongest transitions of $^{172}$Lu, $^{133}$Ba, and $^{152}$Eu in the target. However, the strongest γ 181 keV line of $^{172}$Lu is about 5 times less intense than the γ 574 keV peak of $^{178m2}$Hf. The strongest γ 356 keV line of $^{133}$Ba has approximately the same intensity as the γ 574 keV peak of $^{178m2}$Hf. The intense γ 439 keV line of $^{150}$Eu is around 3 times weaker than the γ 574 keV peak of $^{178m2}$Hf. As a result, the total counting rate for the transitions of all available sources except the $^{178m2}$Hf isomer in the target is less than the total counting rate for the γ 495 keV and γ 574 keV transitions of the $^{178m2}$Hf isomer (in fact the weakest two lines of the $^{178m2}$Hf isomer for which the enhancement has been reported in this work). Thus, in this experiment we used an external source of $^{57}$Co and its strongest transitions with the energy of 122 and 136 keV served as the reference transitions.

The distance between the $^{57}$Co source and detector was that the intensity of the γ 122 keV line in the measured γ-rays spectra is a little stronger than the total counting rate of triggered cascade transitions of the $^{178m2}$Hf isomer. At the same time, the intensity of the γ 136 keV line is somewhat higher than the counting rate of the strongest γ 213 keV peak of the $^{178m2}$Hf isomer. The results for the γ 122 keV reference transition are presented in Fig. 4 demonstrating rather correct statistical distribution. Practically the same result was obtained for the γ 136 keV reference transition.

While the dead time of the data acquisition for the beam-off runs is about 0.4%, during the irradiations at the maximal currents the dead times are less than 0.8%. Since all the measurements have been carried out in the live (not real) time mode the necessary corrections used to be made automatically. Anyway, such small dead times cannot produce any problems in the obtained results. In general, the beam-off and beam-on γ-ray spectra look the same absolutely, except for the low-energy region where the bremsstrahlung peak appears during the irradiations (insert b) in the Fig. 5). None the less, the slight calibration shifts (very small for low-energy peaks, however clearly noticeable for high-energy peaks) and remarkable peak broadenings during the irradiations have been detected (insert a) in the Fig. 5). However, absolutely the same peak broadenings during the irradiations have been detected for the transitions of $^{57}$Co with the energies of 122 keV and 136 keV as well (Fig. 5). The results for both reference transitions do not demonstrate any enhancement and it means that the calibration shifts and peak broadenings during the irradiations cannot influence the obtained results.

It should be underlined that there are no ways to estimate any systematic and non-systematic uncertainties that could be correctly applied to the obtained results. However, the null (within the statistical error) triggering effect in the reference peaks confirms that such uncertainties are insignificant.

At the same time, there could be some concerns that the registered triggering effect can still be produced artificially due to some factors that we have not taken into account or it simply cannot be taken into consideration correctly. One of such factors is a heating of the target and corresponding section of the experimental setup as well. The heating shifts the target a little closer to the detector and that might increase the intensities of registered γ-ray peaks during the irradiations. And although according to our estimates such shift is negligibly small for our previous experiments, signif-
significantly enlarged electron currents in the given experiment could change the situation.

The diameter of our detector is 55 mm and the distance from the detector front face to the detector crystal is 5 mm. Thus, the distance from the detector crystal to the center of the target is $5 + 2 + 2 + (0.3/2) = 9.15$ mm. While the length of the heated target section of the experimental setup is less than 80 mm and its average temperature during the irradiations is less than $200 \, ^\circ\text{C}$, knowing the temperature coefficient of the stainless steel $1.3 \times 10^{-5} \, ^\circ\text{C}^{-1}$ we can estimate the maximal temperature shift of the target as 0.208 mm. In the case of the measurement geometry used during this experiment, such temperature shift could produce no more than 0.8% increase of the detected intensities. In order to check the situation experimentally we repeated the irradiations at distances 19.15 and 29.15 mm as well, i.e. moving the detector by 10 and 20 mm more farther from the target, respectively. To obtain approximately the same statistics, the acquisition time was also increased to 6000 and 8000 seconds instead of the standard 4000 seconds. The results obtained in these triggering experiments are shown by the square symbols with corresponding uncertainties in Fig. 6. The estimated temperature enhancements of the registered intensities are shown by the circle symbols. If we assume that the observed triggering at 9.15 mm is due completely to the thermal effect, the respective temperature enhancements of the registered intensities are expected to be as shown by the triangle symbols. Thus, we can make the conclusion that the heating of the target during the irradiations is not responsible for the observed triggering.

The triggering factor $S$ can be expressed through the triggering cross-section (see also Eq. (4.5) [6] and Eq. (1) [21]) from the relation

$$S(N/\tau) = (N/A) F \sigma_{\text{trig}}$$  \hspace{1cm} (1)

where $N$ is the number of isomeric states in the target, $\tau$ is the lifetime of the isomeric state ($1.4 \times 10^9$ s), $A$ is the area of the target (in units of cm$^2$), $F$ is the flux (current) of incident electrons ($s^{-1}$), and $\sigma_{\text{trig}}$ is the triggering cross-section (cm$^2$) for the isomer de-excitation. $N/\tau$ is the normal decay rate of the isomeric nuclei in the target. Since for the given experiment $N$, $\tau$, $A$ and $\sigma_{\text{trig}}$ are constants, the enhancement factor $S$ turns out to be proportional to the current of incident electrons $F$. Thus, we can combine all the results obtained in this experiment into three groups (Table 2).

First, the results obtained at the electron currents 400–700 μA, second – 750–840 μA and at last – 880–940 μA. The averaged triggering in each group turns out to be $1.29 \pm 0.21$, $1.70 \pm 0.18$ and $1.63 \pm 0.33$, correspondingly. And although the errors are rather high the tendency for the increase of observed triggering at the higher electron currents is quite clear. Having averaged these values one can calculate the total triggering as $1.55 \pm 0.12$ – the same value that was calculated from the data for separate γ-ray peaks analysed in this work and shown in Fig. 3.

3. Discussion

The mechanism of the $^{178m2}$Hf isomer triggering is unclear yet. Within several errors the observed enhancements are the same for the low-lying and high-lying transitions of the $^{178m2}$Hf isomer not
allowing to construct reliably the scheme of $^{178m2}$Hf isomer stimulated decay. If the $^{178m2}$Hf isomer induced decay follows the 13$^+$ excited state of the 8$^-$ band, any intermediate transitions are very low-energy ones. Such low-energy $\gamma$-rays cannot be detected in our experiment. Maybe it is the real reason why no new $\gamma$-ray peaks were recorded in the measured spectra.

The fact that an estimate obtained for the integrated cross-sections of $^{178m2}$Hf triggering is orders of magnitude higher than can be expected for the photon absorption by nuclei in this mass region forced us to assume that atomic processes are of great importance for the triggering process. Thus, in order to explain the observed high efficiency of $^{178m2}$Hf isomer triggering by low-energy photons a number of mechanisms has been proposed. One of such mechanisms is Nuclear Excitation by Electron Transition (NEET), the process inverse to internal electron conversion, when energy of one of the atomic transitions that follow the ionization of atomic shells during the irradiation of $^{178m2}$Hf isomer target by soft X-rays can resonantly be transferred into the nucleus taking simultaneously away some of its quantum momentum and softening greatly the hindrance of corresponding nuclear transition. The NEET effect was studied in detail in $^{197}$Au for which the energy balance between corresponding nuclear and atomic transitions was considered to be as small as 51 ± 2 eV. However, the experiment on NEET observation in $^{197}$Au conducted at SPring-8 [22] has clearly indicated that the energy balance is even less 40 ± 2 eV. Only recently this disagreement was finally resolved [23], confirming reliably the smaller value 39 ± 3 eV.

Another perspective mechanism is Inelastic Electronic Bridge, the process similar to internal Compton effect, when the electron is shaken up to a discrete state, the vacancy in which can be produced during the irradiation of $^{178m2}$Hf sample by soft X-rays, rather than being emitted into the continuum. In this event atomic electrons could take away extra quantum momentum of the nucleus and relax significantly the forbiddenness of corresponding nuclear transition as well [24].

From all the proposed mechanisms of the $^{178m2}$Hf isomer triggering, including the classical NEET, electronic bridges (both elastic and inelastic ones), resonant internal conversion [25] and NEET through autoionization states [26], the classical NEET is considered as the most probable [27]. On the other hand, only this mechanism was tested experimentally and corresponding upper limits for the triggering cross-section have been obtained [6,21]. Therefore, we have also estimated the triggering cross-section that corresponds to the enhancement measured in this work that the triggering is stimulated by photons with the energies higher than the binding energy of $L_3$ electrons in Hf.

The production [28] and absorption of bremsstrahlung photons in the dense Ta matrix was calculated directly by simple integration or when it is not possible – numerically. We assume that the $^{178m2}$Hf isomers are homogeneously distributed over the target thickness. Our analysis was restricted by the photon energies higher than the binding energy of $L_3$ electrons in Hf. As a result, the induced triggering cross-section turns out to be $\sigma_{\text{eh}}^{\text{ph}} = 25$ b. In [21] the upper limit for the magnitude of the photon induced triggering effect cross-section $6.58 \times 10^{-27}$ cm$^2$ keV was obtained and this value is close to the estimate given in [6]. The used photon flux was $2.2 \times 10^{15}$ photons cm$^{-2}$ s$^{-1}$ keV$^{-1}$ or $3.3 \times 10^{12}$ photons cm$^{-2}$ s$^{-1}$, thus the upper limit expressed in barns is 4.4 b. Such value is close to the triggering cross-section obtained in this work.

Although the obtained triggering cross-section is rather high, on the one hand, all our estimates are based on the assumption that the NEET process after the ionization of Hf $L_3$-subshell is responsible for the $^{178m2}$Hf isomer triggering. If the lower energy photons could trigger the $^{178m2}$Hf isomer, then the triggering cross-section would also be remarkably lower.

On the other hand, we should not forget that practically all electron energy goes into the ionization of Ta atoms and only about 0.3% – into the bremsstrahlung radiation. It means that a spot of 1.3 cm diameter and about 2 μm thickness at the target surface presents highly ionized Ta matrix and correspondingly very intense source of Ta characteristic X-rays. And although only 4–5% of Ta characteristic X-rays produced after the ionization of Ta L-subshells have energy high enough to ionize the $L_3$-subshell of Hf, such X-rays (even not so highly directional as the bremsstrahlung radiation, but anyway its one half is still directed into the target) appear to be at least an order of magnitude more intense, than bremsstrahlung photons in the same energy range. Furthermore, the bremsstrahlung photons of all energies produce also the intense ionization of Ta atoms over the target and the corresponding characteristic X-rays of Ta can quite efficiently ionize the $L_3$-subshell of neighboring Hf atoms.

Thus, if all such effects would correctly be taken into account, then the triggering cross-section estimated in the present work could be much less than the upper limit obtained in the earlier papers.

### 4. Conclusions

In summary, we repeated the $^{178m2}$Hf isomer triggering experiment [8] with the experimental setup exploited at the maximally high electron currents reaching 940 μA. The diameter of the beam

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**Table 2**

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<th>3</th>
<th>4</th>
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<th>6</th>
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<td>2.20 (59)</td>
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<td>1.35 (69)</td>
<td>0.26 (60)</td>
<td>0.65 (49)</td>
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<td>1.72 (54)</td>
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<td>2.16 (65)</td>
<td>2.59 (58)</td>
<td>2.37 (76)</td>
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<table>
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<td>1.44 (63)</td>
<td>1.54 (69)</td>
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<tr>
<td><strong>Averaged value for range II + range III</strong></td>
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<tr>
<td><strong>Averaged value for range I + range II + range III</strong></td>
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spot at the target was increased from 8 to 13 mm covering completely all the $^{178}$m$^{2}$Hf isomer activity in the target.

The evidence for the triggering of the $^{178}$m$^{2}$Hf isomer as the enhanced counting rates of all strongest ground-state band (with the energies of 213, 325 and 426 keV) and 8$^{-}$-state band (with the energies of 216, 495 and 574 keV) transitions from the $^{178}$m$^{2}$Hf isomer decay was observed. The total counting rate of above-mentioned cascade transitions was used as a measure of the enhancement factor. The enhancement factor was estimated as 1.55 ± 0.12%. Furthermore, the tendency for the increase of observed triggering at the higher electron currents is quite clear.

In this experiment we used an external source of $^{57}$Co and its strongest transitions with the energy of 122 and 136 keV served as the reference transitions. The results for the γ 122 keV reference transition demonstrate quite correct statistical distribution. Practically the same result was obtained for the γ 136 keV reference transition. It means that at least already known factors such as the slight calibration shifts and peak broadenings during the irradiations cannot influence the obtained results.

The temperature enhancements of the registered intensities were estimated to be negligibly small. Additional triggering experiments conducted at the standard for this work distance from the detector crystal to the center of the target 9.15 mm and the increased distances 19.15 and 29.15 mm show clearly that the heating of the target during the irradiations cannot be responsible for the observed triggering.

While the significances of the obtained triggering effect in the previous $^{178}$m$^{2}$Hf isomer triggering experiments are 3.5 [7] and 4.3 [8] standard deviations, respectively, the significance of the triggering effect obtained in the present experiment is almost 13 standard deviations.

Acknowledgements

The authors are indebted to Prof. Valentine V. Chorny and his colleagues for their help in the development of the experimental setup and to Prof. Alexsey I. Feoktistov and Dr. Vladimir T. Kupryashkin for their help in the various aspects of these measurements. Special thanks to Dr. Nikolay V. Strilchuk for the encouragement of this investigation.

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