Anomalous L-Subshell Internal Conversion of Some Hindered E1 Transitions in ^{171}Tm, ^{175}Lu and ^{177}Hf

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The L-subshell internal conversion electron ratios of some hindered electric dipole transitions in the odd-mass nuclei ^{171}Tm, ^{175}Lu and ^{177} Hf have been measured by means of a high resolution iron-free double focusing beta-ray spectrometer. Large anomalies have been observed in the conversion process. The anomalies are interpreted as an experimental evidence for the presence of dynamic effects. The internal conversion penetration parameters have been determined. The results obtained are discussed in terms of theoretical predictions.

Introduction

The occurrence of so-called anomalous internal conversion can now be used as a means of exploring details of nuclear structure. Deviations from the point-nucleus internal conversion coefficients can occur because of two effects. The first effect we might call static. There the finite radial extension of the central-charge distribution changes the electron wave functions outside the nucleus relative to the point-charge case. A satisfactory account of this effect has changed the assumed values of the theoretical internal conversion coefficient by appreciable amounts [1].

The other effect is sometimes referred to as dynamical and is connected with the penetration of the electron wave function inside the nuclear surface. This penetration usually gives rise to additional nuclear matrix elements not present in the γ-decay. More specifically, the anomaly caused by the penetration depends on the ratio of the nuclear matrix element due to penetration to the normal γ-ray matrix element. It is clear that the effect should be noticeable only if the normal nuclear matrix element is small, i. e. the corresponding γ-transition hindered.

Obviously the probability for the electron to be inside the nucleus is strongly increased with increasing Z and A. The strongest anomalies are found for E1 transitions, and, as expected in line with what has been said above, they occur in the rare earth and actinide region [1] where the volume factor for penetration is most favourable and where the γ-hindrance factors for E1 transitions are particularly large.

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The theory of internal conversion including dynamical nuclear structure effects has been developed by Church and Weneser [2] and by Green and Rose [3]. Special cases (E1 transitions) have been treated by Nilsson and Rasmussen [4]. These authors correlated the observed anomalies, in the conversion coefficients, with the degree of forbiddenness of the gamma-ray transitions and possible model dependent selection rules for the various nuclear matrix elements observed.

As the wave-functions of the K, L_I, L_{II} and L_{III} atomic electrons inside the nucleus differ, the contribution of the penetration terms to the internal conversion process will depend upon the atomic electron being converted. Thus, in cases for which penetration effects are significant, one should expect different degrees of anomaly for the different atomic subshells and hence anomalous subshell conversion line intensity ratios. It was found that for E1 transitions, there are two types of penetration matrix elements, one of these giving an important contribution only to $s_{1/2} \leftrightarrow p_{1/2}$ electron transitions. Such electron transitions are present in L_I and L_{II} conversion, but are absent in L_{III} conversion.

Recently, Pauli [5] studied theoretically the penetration effects on internal conversion coefficients of electric dipole transitions. The conversion coefficient for E1 transitions as given by Pauli [5] is:

$$\alpha = \alpha^{(0)} \cdot \Delta = \alpha^{(0)} (1 + a_1 \eta + a_2 \eta^2 + a_3 \eta \xi + a_4 \xi + a_5 \xi^2) .$$

If the penetration factor Δ has the value one, the conversion coefficient is reduced to its usual value $\alpha^{(0)}$; otherwise the so-called penetration effects are present. The coefficients a_i depend only on the electron wave functions and are to a very high extent independent of the assumptions of nuclear charge distribution and atomic screening. They vary from one subshell to the other, but the nuclear parameters η and ξ are the same for the conversion out of all subshells and can be used as free parameters to fit the internal conversion data. The current parameter η and the charge parameter ξ are defined by Pauli [5].

The anomalous internal conversion data are consistent with the charge parameter $\xi \sim 0$ and one value of the "current parameter η", which exposes furthermore a clear correlation with the hindrance as compared to the Weisskopf estimate [6].

Pauli [5] calculated the current parameter in cases of low energy transitions in an odd-proton and an odd-neutron nuclei, ^{175}Lu and ^{177}Hf respectively. In these nuclei, the three lowest states are interpreted as members of the rotational band built on the ground state, which is a $7/2+$ state in ^{175}Lu and a $7/2-$ state in ^{177}Hf, see Figs. 1 and 2. These levels are fed by E1 transitions from the first excited single-particle state, a $9/2-$ and a $9/2+$ state in ^{175}Lu and ^{177} Hf, respectively.

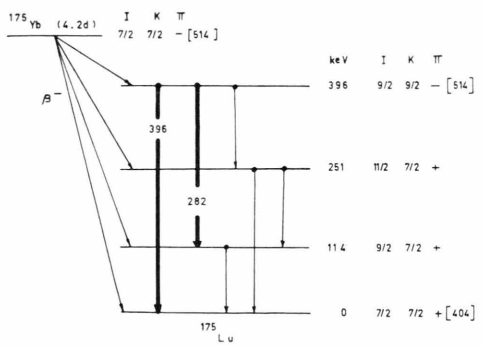

Fig. 1. Level Scheme of ^{175}Lu.

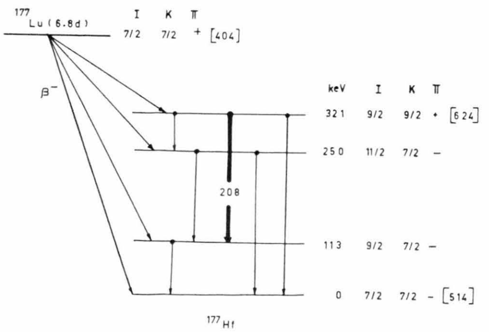

Fig. 2. Level Scheme of ^{177}Hf.

We have previously [7, 8] measured the electron-gamma angular correlations in ^{175}Lu and ^{177}Hf. The angular correlation between the K-conversion electrons of the retarded 208 keV E1 transitions and 113 keV gamma-rays, in ^{177}Hf, was found to be normal whereas the result of the $282\,\mathrm{K} - 114\,\gamma$ angular correlation measurement, in ^{175}Lu, is much smaller than normal. This may be understood in terms of the presence of penetration matrix elements in the conversion process of the retarded 282 keV transition. Later, Holmberg et al. [9] and Thun [10] have remeasured the $(282\,\mathrm{K} - 114\,\gamma)$ angular correlation and they calculated the penetration parameters. There is a discrepancy in their results, and both disagree with the unified-model predictions.

Also, we have measured [11] the K-conversion coefficient of the retarded 321 keV E1 transition in ^{177}Hf, where a large anomaly was observed in the K-conversion process.

Because of these discrepancies and because of the recent interest in the E1 transitions in the rare-earth region, it was apparent that studying the influence of nuclear structure effects on the conversion process of retarded electric dipole transitions in some odd-mass nuclei was desirable, as they may provide strongly model-dependent information on nuclear structure. In these investigations we have measured the L-subshell conversion ratios of the 296 and 308 keV transitions in ^{171}Tm, see Fig. 3, the 396 and 282 keV transitions in ^{177}Lu and finally the 208 keV transition in ^{177}Hf.

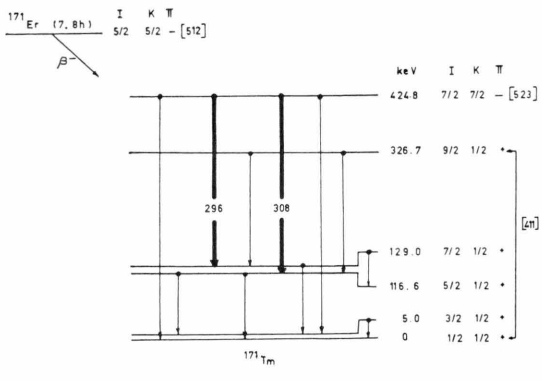

Fig. 3. Low-lying levels of ^{171}Tm.

1. Experiments

1.1. Apparatus

The internal conversion electron data were recorded by means of a high resolution iron-free double focusing beta-ray spectrometer[12] ($\varrho_0 = 50$ cm). By using this instrument relative momentum measurements could be made with an accuracy of a few parts in 10^5. With a 0.2×2 cm^2 source and a 2 mm detector slit a resolution of about 0.15% is obtained. Better resolution could also be obtained for different arrangements[12]. The detector employed in the present studies was a G.M. counter with ~ 1.2 mg/cm^2 mica end window.

1.2. Source preparations

^{171}Er: The radioactive source used in the present work was ^{171}Er which decays by β-decay to ^{171}Tm with a halflife 7.8 h. The sources were prepared by evaporating inactive spectroscopically pure erbium oxide in vacuum onto an aluminium foil of thickness 0.76 mg/cm^2. A special mask was used to obtain samples of dimensions 2×0.2 cm^2. This method was very satisfactory to get a layer of homogeneous activity. The foils were irradiated in a flux of $\sim 10^{12}$ n/cm^2 sec in the ARE-reactor at Inchass. The thickness of the sources ranges between 40–80 μg/cm^2.

^{175}Yb: About 8 mg of natural Yb_2O_3 was placed in a small graphite container and irradiated in the reactor DR2 at Risø, Denmark for 2.5 d at a pile factor of 900. To reduce the handling of the activity, this container was used as an oven in connection with the ion source of an electromagnetic isotope separator. The separation was made at the Department of Physics, Chalmers University of Technology, Gothenburg. By passing CCl_4 through the heated oven, $YbCl_3$ was produced and fed into the ion source. In order to increase the reaction rate, the oxide was mixed with graphite grains. The separation lasted 4 h and the ^{175}Yb activity was collected on an Al-foil having approximate thickness of 1 mg/cm^2. About 2% of the ^{175}Yb activity was collected on the foil and the source obtained in this way had a dimension of 2×0.2 cm^2 and thickness of the order of 20 μg/cm^2.

^{177}Lu: The sources were produced by neutron irradiation of natural spectroscopically pure lutethium oxide in the ARE-reactor at Inchass, over a period of two days in a flux of about 10^{13} n/cm^2 sec. First the Lu_2O_3 was transferred into $LuCl_3$ and then evaporated in vacuum onto an aluminium foil of thickness ~ 1 mg/cm^2. The evaporated material was uniformly distributed in a rectangular form of dimensions 0.15×1.5 cm^2. The thickness of the material deposited was estimated to be ~ 100 μg/cm^2.

1.3. Measurements

The L-subshell conversion line intensity ratios have been determined for highly-retarded E1 transitions in ^{171}Tm, ^{175}Lu and ^{177}Hf nuclei. The conversion electron spectra were studied using the high resolution iron free beta-ray spectrometer. Since most transitions are of fairly high energy, it was necessary to operate at a resolution of $\sim 0.04\%$ in order to resolve the L-subshell lines. The conversion lines were scanned with small and equal current increments. The line intensities were calculated as the sum of counts above background multiplied by the current increment divided by the $B\varrho_0$-value and time used to measure each point. The errors in the electron intensities have been obtained from the expression:

$$N = \sqrt{\Sigma N + 2\,n\,N_{\rm Bgr} + (n\,\Delta N_{\rm Bgr})^2}$$

where ΣN is the sum of counts above background, n is the number of measured points on a line, $N_{\rm Bgr}$ is the number of counts on background and $\Delta N_{\rm Bgr}$ is the uncertainty in the background determination.

The L-subshell ratios were measured for the 296 keV and 308 keV transitions in ^{171}Tm as shown in Figs. 4 and 5. The L-conversion lines of the 282 keV and 396 keV transitions in ^{175}Lu are shown in Figs. 6 and 7. In Fig. 8, we show the experimental results for the 208 keV transition in ^{177}Hf.

Table 1 summarizes the experimental data for the L-subshell ratios. The table also contains the theoretical values of the subshell ratios for E1 transitions obtained from the tabulation of Sliv and Band[13]. The gamma-ray matrix elements $|M|^2$ are quoted relative to Weisskopf units.

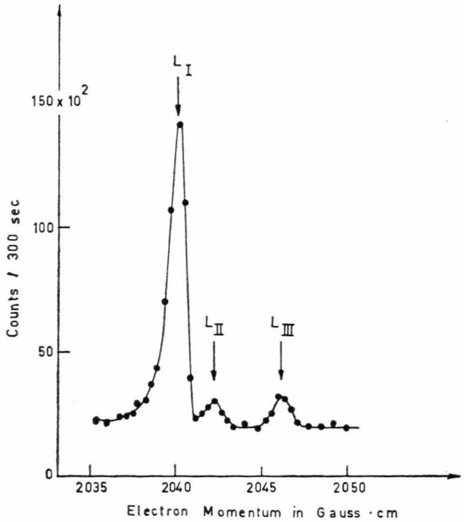

Fig. 4. The L-subshell conversion lines of the 296 keV transition in ^{171}Tm.

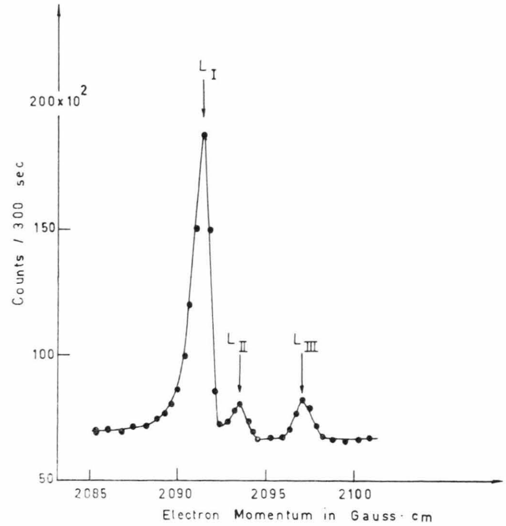

Fig. 5. The L-subshell conversion lines of the 308 keV transition in ^{171}Tm.

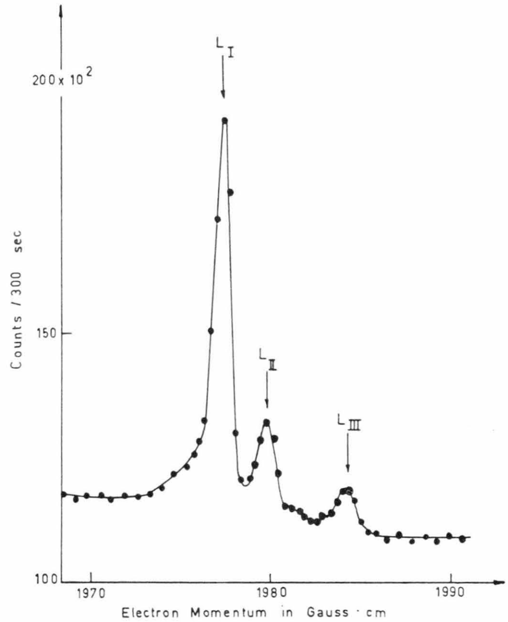

Fig. 6. The L-subshell conversion lines of the 282 keV transition in ^{175}Lu.

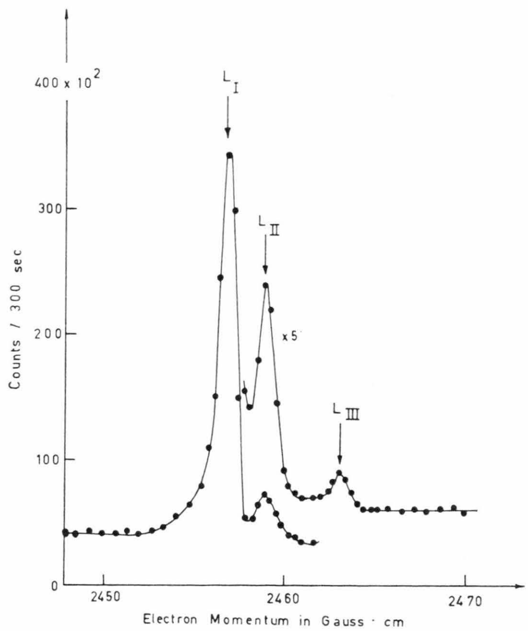

Fig. 7. The L-subshell conversion lines of the 396 keV transition in ^{175}Lu.

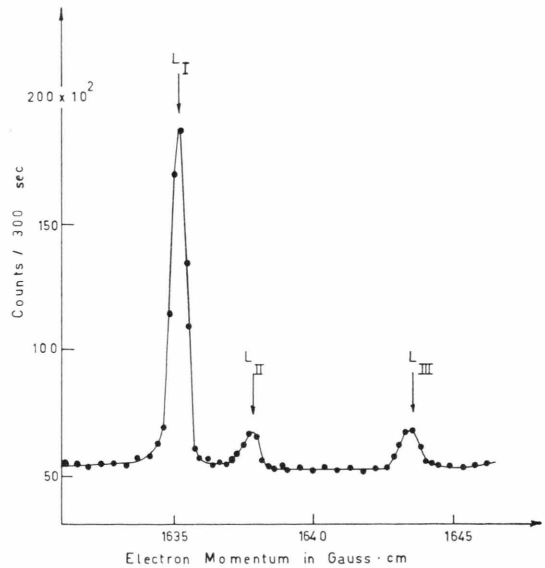

Fig. 8. The L-subshell conversion lines of the 208 keV transition in ^{177}Hf.

2. Results and Discussion

The transitions given in Table 1 have been listed in order of retardation. The most highly inhibited transitions, those in ^{171}Tm, show appreciable anomalies which indicate a penetration contribution to the internal conversion. The experimental ratios are consistent with those quoted recently by Graham et al. [14]. They reported values for $L_I/L_{III} = 10.2$ and 9.0 compared to our results 9.8 and 8.7 for the 296 keV and 308.6 keV transitions respectively. Excellent agreement with the experimental ratios is realized when one includes the effect of the nuclear current penetration term with $\eta = -1.25$ and -2.84 for the 308.6 and 296.0 keV transitions respectively. In making this calculation, it has been

Table 1. L-subshell conversion line intensity ratios and current penetration parameter η for retarded E1 transitions.

Nucleus	E_γ (keV)	$\lvert M\rvert^2$	Theoretical conversion coefficient ratios L_I/L_{III}	L_{II}/L_{III}	Experimental conversion coefficient ratios L_I/L_{III}	L_{II}/L_{III}	Current parameter η
^{171}Tm	296.0	1×10^{-9}	7.7	1.1	9.8 ± 0.8	1.0 ± 0.1	-2.84 ± 0.21
	308.6	2×10^{-9}	7.7	1.1	8.7 ± 0.8	1.12 ± 0.10	-1.25 ± 0.10
^{175}Lu	396.1	6×10^{-7}	9.9	1.1	45 ± 4	5.5 ± 0.5	-13.5 ± 0.5
	282.6	1×10^{-6}	7.6	1.1	12.3 ± 0.9	2.4 ± 0.4	5.8 ± 0.5
^{177}Hf	208.4	3×10^{-5}	5.8	0.98	6.2 ± 0.5	1.0 ± 0.1	-0.4 ± 0.3

assumed that the influence of the nuclear charge parameter is negligible.

The subshell ratios for the transitions in ^{175}Lu are very different from the theoretical values, see Table 1. In our analysis we have assumed that the L_{III} conversion coefficient is not affected by the penetration effects, and we have used the experimental L_{III} conversion coefficients ($\alpha_{L_{III}}$) to determine the M2 admixture in the E1 transition. We have used the published [7] values of α_K and our measured ratios of K/L_{III} to determine the values of L_{III}. For the transitions in ^{175}Lu these values show that the M2 admixture is less than 3% in the two cases. For the 282.6 keV transition, $\gamma-\gamma$ angular correlation [7] studies have shown that the admixture is less than 1%. Such small M2 admixtures do not significantly effect the L-subshell ratios. The anomaly factor tends to decrease with decreasing retardation but is not directly proportional to the γ-ray retardation. In case of the 396 keV transition, the current penetration parameter was calculated to be -13.5 ± 0.5 which is compatible with that calculated by Pauli [5] (-13.0) using the K and L subshell conversion coefficients which have been measured by Hager and Seltzer [15] and by Emery and Perlman [16].

The penetration contributions in the internal conversion process of the 282.6 keV transition have been deduced [9] by use of measured K-electron particle parameters and the K-conversion coefficients obtained by other groups. The penetration parameter obtained [9] is fairly consistent with our result. A measurement of the particle parameter would give also the conclusive information in this case. This particle parameter cannot be determined in a usual angular correlation experiment, but in a nuclear alignment experiment where the angular distribution of the conversion electrons was measured. Thun [10] has re-measured the $(282\,K-114\,\gamma)$ correlation and re-evaluated the penetration parameters. The discrepancy between Thun's [10] and Holmberg's et al. [9] results seems to stem from a difference in the formulae used for the mixed

Previously, we have measured [8] gamma-gamma, particle parameter.
electron-gamma and gamma-electron angular correlations of the 208 keV – 113 keV cascade in ^{177}Hf. The angular correlation between the K-conversion electrons of the retarded 208 keV E1 transition and 113 keV gamma-rays was found to be normal. Recently, the internal conversion process of the 208 keV retarded E1 transition was studied [17], by measuring the electron-gamma and γ-γ directional correlations of the 208 – 113 keV cascade. The result obtained for the ratio between directional correlation coefficients -1.66 ± 0.04 is in agreement with the theoretical value for a pure E1 transition -1.68. The result of the nuclear current penetration parameter -0.15 ± 0.25, is found to be in agreement with our present result -0.4 ± 0.3 and is also consistent with that calculated from the penetration effect of the 321 keV transition in ^{177}Hf by use of the branching rules in the rotational model.

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Notizen

Assoziationskontinua der positiven Molekülionen im Kern von Niederstromlichtbögen

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Association Continua of the Positive Molecular Ions in the Core of Low Current Arcs

Measurements on low current Cl_2-, Br_2-, J_2-, and $AlCl_3$-arcs show an intense continuous radiation in the visible and near infra-red spectral range, which is interpreted as association continuum of the various molecular ions.

Einleitung

Bei quantitativen Messungen an Niederstrombögen zur Bestimmung der „Detachment-Querschnitte" von negativen Ionen wurde von Mück und Popp [1] im Chlor-, von Frank, Neiger und Popp [2] im Brom- und von Neiger [3] im Jodplasma neben der isoliert auftretenden Strahlung des jeweiligen Affinitätskontinuums (freigebunden-minus-Strahlung) im UV-Spektralbereich stets eine intensive, nahezu weiße Untergrundstrahlung festgestellt.

Nach Abzug der entsprechenden Brems- und Rekombinations-Strahlungsanteile der Elektronen und positiven Ionen (frei-frei- und frei-gebunden-Strahlung), die mit Hilfe der Theorie von Kramers und Unsöld [4] berechnet wurden, verbleibt in allen drei Fällen eine beträchtliche „Reststrahlung". Der Temperaturgang dieser „Reststrahlung" ist in erster Näherung proportional dem jeweiligen Produkt aus der Neutralteilchendichte N_0 und der Elektronenteilchendichte N_e. Auf Grund dieser Tatsache erschien es naheliegend, die erhaltene „Reststrahlung" als Bremsstrahlung der Elektronen im Felde der neutralen Atome (frei-frei-minus-Strahlung) zu interpretieren, zumal auch die Wellenlängenabhängigkeit der des frei-frei-minus-Kontinuums entsprach.

Sonderdruckanforderungen an Dr. H.-P. Popp, OSRAM GmbH-Forschung, *D-8000 München 90*, Hellabrunner Straße 1.

Im Falle des Chlorplasmas [1] liefert jedoch die spezielle Theorie der frei-frei-minus-Strahlung von Kandel [5] Werte, die um etwa den Faktor 200 zu klein sind. Für das Brom- [2] und Jodplasma [3] ergibt sich die Theorie von Kas'yanov und Starostin [6] Abweichungen um etwa zwei Zehnerpotenzen von den erhaltenen „Restkontinua".

Neue Messungen an einem reinen $AlCl_3$-Niederstromlichtbogen bei Atmosphärendruck [7] zeigen wiederum ein intensives „Restkontinuum" im sichtbaren und IR-Spektralbereich, dessen Temperatur- und Wellenlängenabhängigkeit wie in den Fällen des Chlor-, Brom- und Jodplasmas auf einen wirksamen frei-frei-minus-Strahlungsmechanismus hinweisen. Doch auch beim $AlCl_3$-Plasma liegen die theoretisch berechneten Werte um ca. zwei Zehnerpotenzen unter den Werten des Experiments.

Die Theorie von Kas'yanov und Starostin [6] ist inzwischen in einer Reihe von Arbeiten verschiedener Autoren auf Glimmentladungen [8, 9], Bogen- [10] und Stoßwellenplasma [11] angewendet und bestätigt worden. Neuere Theorien von Mjolsness und Ruppel [12], Geltman [13] sowie Hyman und Kivel [14] zur Beschreibung der frei-frei-minus-Strahlung liefern ähnliche Ergebnisse. Die Abweichungen der einzelnen Theorien voneinander betragen in der Regel maximal den Faktor 5.

Aus diesen Gründen erscheint die Theorie von Kas'yanov und Starostin [6] weitgehend gesichert. Berechnet man nun mit Hilfe dieser Theorie die für die Bremsstrahlung der Elektronen im Felde der neutralen Chloratome wirksamen Streuquerschnitte einerseits aus den Ergebnissen am reinen Chlorplasma [1] und andererseits aus den Ergebnissen am $AlCl_3$-Experiment [7], so liefert sie Werte, die bis zu einer Zehnerpotenz voneinander abweichen.