# Upper-limit determination of resonant trielectronic recombination cross-section for krypton using crystal channeling

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We used channeling through a thin crystal to get an estimate of the cross section of the resonant trielectronic capture by $Kr^{34+}$ ions. *K* x-ray–*K* x-ray coincidence measurements were performed with a selection on the charge state and energy loss of transmitted ions. An upper limit of $1.9\times10^{-27}$ cm$^2$ at the resonance is obtained, and this represents an improvement by two orders of magnitude with respect to previous ion-gas experiments. The possibility to reach the theoretical predictions experimentally is discussed.

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## I. INTRODUCTION

Trielectronic recombination (TR) is a resonant electron capture process that may occur during heavy ion-electron collisions: the capture of an electron by an ion with at least two electrons in the initial state is accompanied by the simultaneous double electronic excitation of the ion. This process is due to a three-electron interaction that is analogous to dielectronic recombination (DR), in which electron capture is accompanied by a single excitation. Figure 1 presents these electronic transitions in the ion rest frame. Figure 1(a) illustrates the particular case of the *KK-LLL* resonant trielectronic recombination for a He-like ion: the energy gained during the capture into the $n=2$ state of an electron with the required kinetic energy in the ion frame is devoted to the $(1s^2\rightarrow2\ell2\ell')$ excitation of the two *K* electrons. This leads to an excited Li-like ion with its three electrons in the *L* shell (''triply'' excited). For sufficiently high *Z* ions, the radiative decay of this triply excited state by two successive *K* x-ray emissions dominates autoionization, and recombination effectively occurs. The analogy with DR is shown in Fig. 1(b) for the *K-LL* resonance, occurring at a lower electron kinetic energy.

Of course, resonant electron-capture processes similar to DR and TR can take place during ion-atom collisions. These processes are called respectively resonant transfer and excitation (RTE) and, following Zaharakis *et al.* [1], resonant transfer and double excitation (RT2E). RTE is the capture of an initially bound electron, and can lead either to autoionization (RTEA) or radiative stabilization (RTEX). An important feature of RTE (and RT2E), if one compares them to DR (and TR), is the broadening of the resonance due to the Compton profile of the electron to be captured, i.e., the longitudinal momentum distribution in the initial state. For the corresponding trielectronic process, we may use RT2E2A, RT2EAX, and RT2E2X to denote two-Auger, Auger +radiative, and double radiative decay of the intermediate state, respectively.

For low *Z* ions, the triply excited state formed during trielectronic recombination decays mostly by autoionization. This has been observed for the lightest ions: Schultz [2] reported on resonances above the ionization threshold produced during electron scattering or electron-impact ionization experiments that originate from the formation of $2\ell^2 2\ell'$ states of $H^{2-}$ and $He^-$ ions. More recently, Müller *et al.* [3] measured the cross sections for the resonant capture into $n=2$ triply excited states of Li atoms by means of very precise $Li^+$-electron crossed-beam experiments, by using the energy dependence of electron-impact ionization: with a resonance strength of about $10^{-20}$ cm$^2$ eV, this process (which keeps the ion charge constant) lowers the electron-impact ionization of $Li^+$ ions significantly (by less than 1%).

The time reversal process of the resonant capture during trielectronic recombination (trielectronic capture) is the three-electron Auger decay, also called the ''double'' Auger effect [DA, double deexcitation–single ionization; see Fig. 1(c)], that has been observed in ion-atom [4,5] and ion-surface [6,7] collision experiments. Excited light ions with a double *K* hole were produced [5–7], and the ratios *R* of DA to single Auger rates for such states were found to be around $3\times10^{-4}$ for C and N ions [5,7]. Following the detailed balance principle, these ratios should be similar to the ratios of trielectronic capture over dielectronic capture cross sections.

Trielectronic capture—and DA—are of fundamental interest for the study of three-electron correlation in bound systems. Most of the theoretical works on DA have con-

 

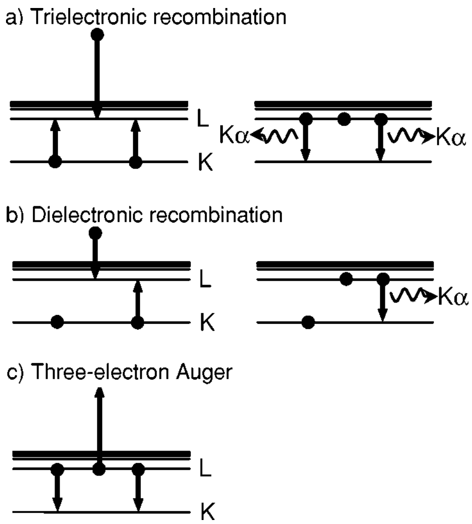


FIG. 1. Schematic representations of (a) *KK-LLL* trielectronic recombination (TR) for an initial ground-state He-like ion (b) *K-LL* dielectronic recombination (DR) for the same initial state (c) *KK-LLL* three-electron Auger (DA).

cerned low-$Z$ ions [8,9]. Vaeck and Hansen [9] pointed out that, for double-$K$-hole nitrogen ions, the main contribution to DA comes from the shake-down mechanism accompanying a single autoionization, when at least one $2s$ electron is present in the initial state. A shake-down transition can take place because the initial $2s$ and the final $1s$ wave functions are not orthogonal. Their calculations show that $R$ values depend more strongly on the number of $2s$ electrons than on the total number of $L$ electrons. Thus ''real correlation'' effects (e.g., those involving only $2p$ electrons) lead to values of $R$ much smaller than those involving initial $2s$ electrons (typically $\sim 10^{-6}$ and $\sim 10^{-4}$, respectively; this last number being in agreement with experimental results [7]). Marques *et al.* [10] used multiconfiguration Dirac-Fock (MCDF) calculations to estimate DA rates and the ratio $R$ for higher $Z$ systems ($Z=36,41,64$). They did not include the shake-down contribution, which was assumed to be small ($\sim$20% of the total DA rate for krypton, and less for higher $Z$ ions). Their final calculations provided a value of $R=2.786 \times 10^{-7}$ for triply excited $Kr^{33+}$ (three electrons in the $L$ shell). For the same ion Badnell [1,11] estimated a ratio $R \sim 10^{-6}$ by using a three-$L$-electron interaction through configuration mixing with a Breit-Pauli Hamiltonian.

For those high-$Z$ ions, the experimental study of trielectronic recombination is more attractive than the study of DA, because the radiative decay of a double $K$ hole dominates autoionization. Using an initial He-like ion in its ground state, the signature of the *KK-LLL* trielectronic recombination is a double $K$ x-ray emission, along with the decrease of the ion charge by one unit.

One attempt to observe RT2E is reported by Zaharakis *et al.* in Ref. [1]. During $Kr^{34+}$-ion–$H_2$-gas collisions, they detected x rays in coincidence with $Kr^{33+}$ transmitted ions at various incidence energies. Due to the low interaction rates in such experiments, they could only measure the energy dependence of the sum of the $L,M,\ldots$ REC cross sections ($\sim 10^{-23}$ cm$^2$), and no $K\alpha$ photon could be detected in coincidence. They gave an upper limit of the RT2E cross section of about $10^{-25}$ cm$^2$.

Crystal channeling provides a powerful technique for investigating such a low-cross-section process, because channeled ions interact mainly with a dense gas of quasifree electrons. In alignment conditions, ions acquire a transverse energy at the entrance of the crystal, according to their incidence angle and their position relative to the atomic rows or planes. Ions with a transverse energy (per unit charge) below the maximum value of the continuum string or planar crystal potential are known to be channeled and can access only a restricted part of the transverse space [12]. This results in the extinction of close collisions with target atomic cores, and the attendant reduction of ion-electron interactions, such as energy loss. One can then study ion-electron interaction processes that are hardly observable in classical ion-solid or ion-atom collisions, as was demonstrated in previous studies of radiative electron capture (REC) [13–15], electron-impact ionization and excitation (EII and EIE, respectively) [16–18], and resonant transfer and excitation [19–21]. A review of charge-exchange experiments in channeling conditions is given in Ref. [22]. The processes just mentioned have relatively high cross sections (more than 10 b), and thus are easily observable in channeling conditions. For instance, the impact-parameter dependence of REC line shapes was studied in detail [15], and the Compton profiles of silicon valence electrons were found to be very close to those of free electrons (Fermi gas) for the same local density. These Compton profiles are also responsible for the broadening of the RTE resonance profiles reported in Refs. [19–21]. The conditions for observing the very rare TR process in channeling conditions are more stringent than those discussed above. They are described in the following sections.

## II. EXPERIMENT

A primary beam of 43.1-$A$ MeV $Kr^{31+}$ from the GANIL facility (Caen, France) was used in our study. The final beam on the target was obtained by means of degrading/stripping Al foils followed by a magnetic spectrometer for the selection of the charge (34+), the energy (37.1, 40.6, and 42.7 $A$ MeV with $\Delta p/p=10^{-4}$), and the emittance ($\epsilon_x=\epsilon_y=0.15\pi$ mm mrad). The two extreme incidence energies are outside the *KK-LLL* resonance, whereas 40.6 $A$ MeV is on the resonance. The beam, with a typical intensity of $(2-3)\times 10^6$ ions/s (within a 9.4-MHz HF pulsed structure) was sent to the Spectromètre à Perte d'Énergie pour le GANIL (SPEG) [23] beam line, and had a diameter of 1 mm on the target. A sketch of the experimental setup in the SPEG line is shown in Fig. 2. This beam line is equipped with a high-resolution spectrometer (8.1 mm on the focal plane corresponding to $\Delta p/p=10^{-3}$). The target was a 3.6-$\mu$m-thick Si crystal oriented along the $\langle 110\rangle$ axial direction. A high efficiency for the detection of projectile $K$ x-ray–$K$ x-ray coincidences was achieved by means of three large NaI(Tl) detectors covering 25% of the total solid angle. These detectors were chosen for their large area and relatively high counting rate capabilities, despite their poor energy resolution. Two

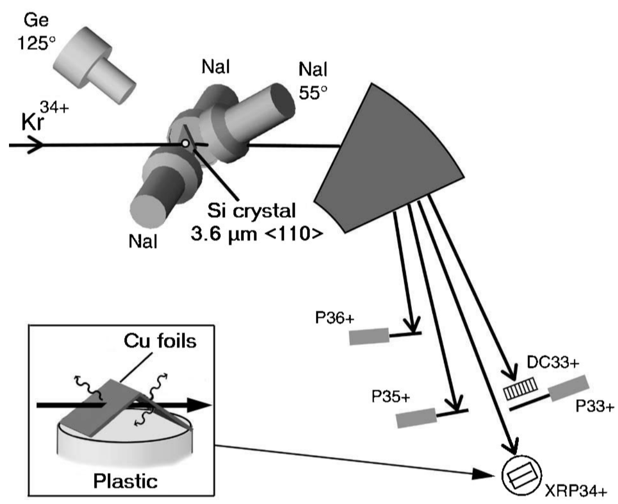


FIG. 2. A schematic of the experimental setup.

detectors were placed at 90° from the beam direction, and the third one at 55°. The $K$ x-ray–$K$ x-ray coincidence efficiency was $\epsilon_{KK}=3.7\pm0.4\%$ (assuming an isotropic emission in the projectile frame). An additional Ge detector at 125° with a much smaller solid angle was used for observing x rays with a better energy resolution. Transmitted ions were charge- and energy-analyzed by the last magnetic stage of the SPEG spectrometer. Down-charged 33+ ions were detected by a drift chamber (DC33+) for position information and a plastic scintillator (P33+) for triggering (see Fig. 2). The main trigger of the event-by-event acquisition (i.e., the trigger for possible TR events) was the detection of a 33+ ion in coincidence with two photons registered simultaneously in two NaI(Tl) detectors. Additional triggers [single events registered by the germanium, NaI(Tl) and DC33+ detectors] were used to observe single charge-exchange and excitation processes.

The fraction of accelerator-HF pulses containing more than one incident ion was not negligible. The probability of undue triggering (detection of three uncorrelated events) increases as the cube of the number of ions per pulse, and then the experimental background increases correspondingly. Thus we detected all the transmitted ions in such a way that we could select, off line, those events for which no 34+, 35+, or 36+ ion were detected within the same pulse as the 33+ ion. During preliminary tests, the plastic scintillators (type BC400) were found to detect up to $\sim10^8$ Kr ions per $mm^2$ in our energy range before the reduction of their efficiency by radiation damage became significant. In the present experiment the charge-state distribution in channeling conditions (slightly varying with the incident energy) is $F(36+)\leqslant1\%$, $F(35+)\simeq3.5\%$, $F(34+)\simeq95\%$, $F(33+)\leqslant1\%$, $F(32+)$ being negligible. The 36+ and 35+ ions were detected by plastic scintillators (P36+ and P35+ respectively; see Fig. 2). The location of the beam impact on these scintillators was changed when necessary. The detection of 34+ ions required a special arrangement because of its very-high-counting rate. The detector (XRP34+) was based on x-ray fluorescence. Copper radiator foils were placed on the top of a plastic scintillator (see the inset of Fig. 2). A very large number of simultaneous x rays were emitted inside the radiator (mainly Cu $K\alpha$ arising from Cu atom ionization). This led to a measurable signal in the large solid angle scintillator with a 100% efficiency.

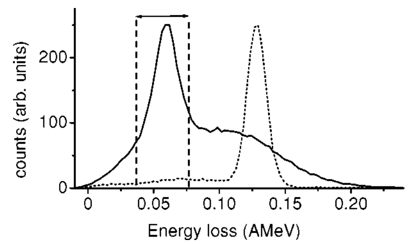


FIG. 3. Energy-loss distributions for axial (full line) and random (dashed line) crystal orientations. The energy-loss range (axial alignment) selected in the data analysis is indicated.

### III. RESULTS AND ANALYSIS

Channeled ions are characterized by their low-energy loss rate. Figure 3 presents two energy-loss distributions for transmitted 33+ ions as measured by the drift chamber at 37.1-$A$ MeV incidence energy. One corresponds to a random crystal orientation and the other one is obtained in axial channeling conditions ($\langle110\rangle$ orientation). The width of the ''random'' distribution is dominated by energy straggling (the width of the direct beam is typically 50% narrower and is mainly due to the size of the beam spot on the target). A low-energy loss tail (extending down to 0) is due to etch pits in the crystal. These pits were formed during the chemical etching of the crystal. The spectrum obtained in alignment conditions has two distinct components: a narrow distribution, with a most probable energy loss roughly equal to half of the random energy loss, and a broader one, centered at a higher energy loss, that corresponds to unchanneled ions. In principle, making cuts in the energy-loss spectrum allows one to select ions with a given transverse energy inside the crystal channel. This transverse energy is related to the mean electronic density encountered inside the crystal channel (see, for instance, Refs. [22,21,17,15]). Our aim was initially to study RT2E as a function of the sampled electron density. The RT2E signal results from a triple coincidence of $K$ x-ray–$K$ x-ray–$Kr^{33+}$ detection arising from a single collisional event. The background signals have the same signature but originate from three uncorrelated events during the ion's passage through the crystal, because $K$ emission cannot result from a nonresonant capture by a He-like ion. Hence it is proportional to the cube of the number of target electrons. Then the ratio of the RT2E signal to the background rate varies as the inverse square of the electron target ''thickness.'' Unexpectedly this background was observed to increase for the lowest-energy losses. Two factors can account for this. First the total thickness of the amorphous layers ($SiO_2$) on the two surfaces of the crystal was measured to be 100 Å (equivalent Si thickness for the RBS technique used). Mechanical electron capture (MEC) and nuclear impact ionization (NIE) can occur in these layers, whatever the transverse energy of channeled ions is. Both MEC and NIE have much larger cross sections than REC and EIE, which are the

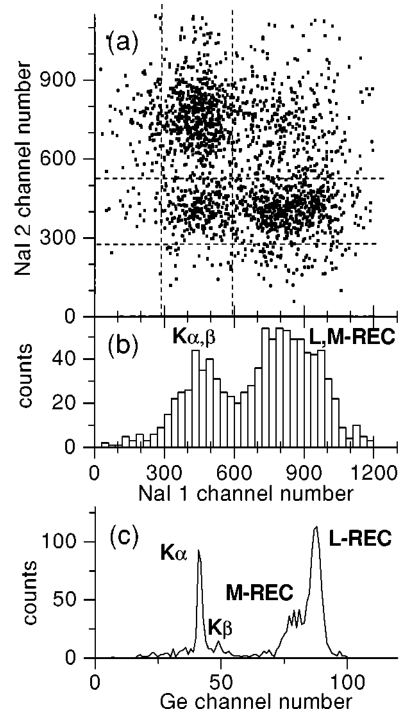


FIG. 4. (a) Scatter plot of the coincidence x-ray–x-ray distribution for two NaI(Tl) detectors (located at 90° from the beam direction), in coincidence with $Kr^{33+}$ ions with the selected energy loss. (b) Histogram of the NaI1 events corresponding to the $K$ x ray on NaI2 for the energy cut shown in (a). (c) Energy spectrum of the Ge detector (125°) for x rays in coincidence with $Kr^{33+}$ ions.

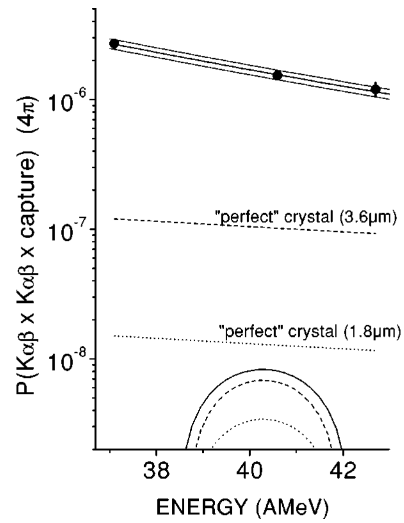


FIG. 5. Probability, for ions in the selected energy-loss range, of emitting two $K\alpha\beta$ photons and capturing one electron in the crystal. The lines are the result of a fit (weighted by the experimental error bars) with $\pm 2\sigma$ standard deviations. The lower solid curve is the expected RT2E resonance profile for channeled ions sampling a density of $0.2e^{-}$ Å$^{-3}$. Dashed curves: calculated background and resonance profiles for a ''perfect'' crystal (3.6 $\mu$m thick, sampled electron density: $0.15e^{-}$ Å$^{3}$). Dotted curves: the same, for a 1.8-$\mu$m-thick ''perfect'' crystal.

only processes likely to occur during ion-electron collisions in channeling. Second, the nonhomogeneity of the target thickness mentioned above can allow a small part of nonchanneled ions to lose very little energy in thinner parts of the target, even less than channeled ions sampling the full crystal thickness. Thus, we decided to optimize the fraction of real channeled ions by selecting particles having their energy loss between 0.45 and 0.6 times the normal energy loss (random orientation), as indicated by the two vertical lines in Fig. 3. No severe selection in transverse energy was made, and the mean sampled electron density was estimated to be equal to the total valence electron density in silicon, i.e., $0.2e^{-}$ Å$^{-3}$. This electron density was deduced with an accuracy of $\pm 20\%$ from the amplitude of $L,M$ REC lines in the x-ray spectra, the cross sections of which are well known [24].

Sample x-ray spectra are presented in Fig. 4. In Fig. 4(a) we show one of the NaI(Tl)-NaI(Tl) two-dimensional x-ray coincidence spectra recorded at a beam energy of 37.1 $A$ MeV, after selection of the 33+ ions as discussed above. Cuts were made in these spectra, as represented in Fig. 4(b) [histogram of the NaI1 events after selection of $K$ x rays registered in NaI2, as shown in Fig. 4(a)]. Figures 4(b) and 4(c) allow the comparison between such a triple coincidence NaI(Tl) spectrum and a spectrum obtained with the Ge detector (in coincidence with 33+ ions). Due to a much better energy resolution of the Ge detector, one clearly sees $K\alpha,\beta$ components, and $L,M,\ldots$ REC lines at higher energies. A small bremsstrahlung contribution is also visible around the $K$ x rays. The NaI(Tl) detectors are only able to resolve the $K$ x-ray peak (that contains also some bremsstrahlung) and the REC peak. Note that the relative amplitudes between $K$ x rays and REC on the two spectra are different because the observation angles are different and also because the NaI(Tl) spectrum is conditioned by the detection of a second $K$ x ray.

## IV. DISCUSSION

The probability for a channeled ion (actually, an ion selected in the energy-loss range defined above) to emit two $K$ x rays in the full solid angle and to emerge from the crystal as 33+ is represented in Fig. 5 at the three incidence energies. The error bars are due to statistics. No evidence for a resonance is found at 40.6 $A$ MeV. The solid line through the experimental points is the result from a fit of these data [with a function $P(E)=aE^{-6}$]. The power $(-6)$ of the energy dependence, used in the fitting function, reflects the dominance of MEC and NIE over REC and EIE. Indeed, $P$(MEC) varies with energy as $E^{-5.5}$, $P(L-\text{REC})$ as $E^{-1.8}$, and $P$(NIE) and $P$(EIE) depend little on the incident energy [25]. This shows that crystal defects are the main source of background in this experiment: for a perfect crystal, one would have had only the background due to REC-EIE-EIE sequential events, with an energy dependence close to $E^{-1.8}$.

The lines above and below the best fit correspond to $\pm 2\sigma$ deviations of the fitting parameter *a*. From this fit one can extract an upper limit of the trielectronic recombination cross section per target electron at the resonance energy. With a 95% confidence level (based on statistics) we obtain $\sigma(\text{RT2E})<1.9\times 10^{-27}$ cm$^2$. Systematic uncertainties of

30% on this value are due to the $K$ x-ray–$K$ x-ray coincidence absolute efficiency ($\sim\pm10\%$), the absolute dose determination ($\sim\pm20\%$, that comes from the photomultiplier background on the 34+ detector at high counting rates), and to the mean target electron density estimate ($\sim\pm20\%$).

The expected shape of the trielectronic recombination resonance is also shown in Fig. 5. We used here the Compton profile of a free-electron gas with a density $\rho_e=0.2e^-$ Å$^{-3}$, as we did in previous RTE studies [20]. The amplitude of the calculated resonance was deduced from the final calculations of Marques *et al.* [10]. These authors estimated the ratio $R$ of DA over single Auger rates for $LLL$ triply excited states to be $3\times10^{-7}$ for Kr ions. This ratio is close to the ratio of trielectronic over dielectronic recombination rates, or for RT2E over RTE cross sections (using the same electron Compton profile). Badnell calculated the ratio of DA over single-electron Auger rates to be somewhat higher ($R\sim10^{-6}$ [1,11]), and also made calculations for RTE for $Kr^{35+}$ ions incident on $H_2$ targets [26]. We can conclude from our experimental results that $R$ is smaller than $5\times10^{-6}$. Considering reasonably that the sensitivity of the experiment reported in Ref. [1] is limited to $\sim$10% of the $(L+M)$-REC cross sections (from their error bars), i.e., to about $10^{-24}$ cm$^2$ per target electron, our results represent an improvement by more than two orders of magnitude for the upper limit of the trielectronic recombination cross section. However, using the same technique (triple coincidences in channeling conditions), one could increase the sensitivity of our experiment by more than one order of magnitude, and reach the range of the theoretical estimates, by the following considerations: results of calculations of the background probability resulting from pure ion-electron interaction (i.e., $L,M,\ldots$ REC–K-L EIE–K-L EIE) are given in Fig. 5 (dashed lines): they correspond to a ''perfect'' crystal, i.e., with an homogeneous thickness and without amorphous layers. Using such a crystal would allow one to select lower electron densities for well channeled ions (around $0.15e^-$ Å$^{-3}$, as we did in a previous REC experiment [15]). For the same crystal thickness this background is calculated to be below $10^{-7}$ at the resonance energy. Note that the corresponding calculated resonance profile is narrower, because both the width and the amplitude depend on the electron density. Since the probability of these three uncorrelated events varies as the cube of the target thickness, using a thinner crystal would certainly make the trielectronic recombination cross sections reachable: we present also in Fig. 5 the calculations for a 1.8-$\mu$m-thick ''perfect'' crystal (dotted line), for which the signal to background ratio would be enhanced by another factor of 4 as compared to the 3.6-$\mu$m-thick crystal. These last calculations correspond to the almost thinnest crystals that can realistically be used with our experimental setup (energy losses have to be resolved between channeled and unchanneled ions). We should mention at this point that the available theoretical estimates of DA rates at the time of our experimental run were higher by one order of magnitude than those published in Ref. [10]. For comparison, the same signal to background ratio would be obtained for a 1.8-$\mu$m-thick ''perfect'' crystal or for a 7.5-$\mu$g/cm$^2$ amorphous carbon target (the MEC-NIE-NIE background is considerably lower for low-$Z$ targets). However, in the latter case, the RT2E rate would be lower by one order of magnitude, which would make the accumulation of statistics out of reach within reasonable beam times.

## V. SUMMARY AND CONCLUSION

In summary, crystal channeling and triple coincidence measurements ($K$ x-ray–$K$ x-ray–$e^-$ capture by a channeled ion) was used to estimate the $KK$-$LLL$ trielectronic recombination cross section for He-like krypton ions. An upper limit is obtained that corresponds to an improvement by two orders of magnitude over that obtained in previous ion-gas experiments. The present result is compatible with theoretical predictions: the shake-down mechanism that provides the main contribution to double Auger rates for low-$Z$ systems has a much smaller contribution at $Z=36$ (the shake-up, corresponding to trielectronic recombination, is not measurable here). The ''pure'' three-electron correlation contribution to DA (and TR) is more than one order of magnitude below the sensitivity of our experiment. The use of much better quality crystals could certainly help to observe this exotic electron capture mode.

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