Single Transfer-Excitation Resonance Observed via the Two-Photon Decay in He-like Ge^{30+}

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We measured the $2E1$ decay of the $1s2s\,{}^1S_0 \rightarrow 1s^2\,{}^1S_0$ transition in He-like germanium for 12- to 19-MeV/u $Ge^{31+} + H_2$ collisions. The resonant population of the $2s2p\,{}^1P_1$ state by transfer excitation was isolated due to its cascading to the $1s2s\,{}^1S_0$ state. The experimental cross sections compare well with calculations using dielectronic recombination rates. The method gives the unique possibility to populate selectively the ${}^1S_0$ state in heavy He-like ions.

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During recent years much effort has gone into the investigation of dielectronic recombination (DR).[1] In an ion-electron encounter a free electron is captured and a bound electron is excited resonantly by electron-electron interaction resulting, generally, in a doubly excited intermediate state of the charge-changed ion, $(q-1)^+$, which subsequently stabilizes radiatively. Dielectronic capture, the first step in DR, can be treated as the time reversal of the Auger effect.[2] A corresponding process to DR in ion-atom collisions is resonant transfer and excitation (RTE).[3-5] However, the momentum distribution of the bound target electrons—described by the Compton profile—leads to a considerable broadening of the resonances, in particular, for light projectiles. Even in the most fortunate cases of inner-shell excitation (e.g., *KLL* resonances, where the Auger notation is used) for the heaviest few-electron projectiles, a separation of neighboring resonances is not directly possible[6] as long as we consider the charge-conserving decay channel for the doubly excited states (for autoionizing channels, see, e.g., Ref. 7).

RTE as well as DR have both been advocated as methods to populate selectively certain doubly excited atomic states normally not accessible by other methods. Even though RTE investigations have the important experimental advantages of a high density of the atomic-confined target electrons and of an interaction region easily accessible for a radiation spectroscopy, it seemed that the broad Compton profile would prevent the isolation of a single resonance. In this Letter we report a method to separate one single resonance in RTE collisions for hydrogenic heavy projectiles leading by radiative stabilization to a dominant population of the metastable state in He-like ions which decays only by the rare $2E1$ transition.

For hydrogenic projectiles the doubly excited intermediate states produced by inner-shell RTE all have two K vacancies, resulting in the emission of two coincident K x rays. Therefore, for hydrogenic projectiles the x-ray–x-ray coincidence technique is suitable to observe the decay of *KLn* resonances, which has already been demonstrated for RTE in S^{15+} on H_2 collisions.[8] Among the different overlapping *KLL* RTE resonances in He-like ions there is one leading to the doubly excited $2s2p\,{}^1P_1$ state which decays promptly into the $1s2s\,{}^1S_0$ state and this can only decay via a two-photon decay $(2E1)$ into the $1s^2\,{}^1S_0$ ground state. As the $2s2p\,{}^1P_1$ state is the only dominant one feeding the metastable $1s2s\,{}^1S_0$ state at *KLL* resonances, the $2E1$ decay of this state provides a means to isolate the RTE resonance via the $2s2p\,{}^1P_1$ level for heavy projectiles. For heavy He-like ions the $2E1$ decay is already fast enough [transition rate $\propto Z^6$ (Ref. 9)] for detection near the reaction volume. In the decay of the $2s2p\,{}^1P_1$ state three x rays are emitted: one K x ray from the decay into the $1s2s\,{}^1S_0$ state and two photons of continuous energy, where the sum of both is the $1s2s\,{}^1S_0$–$1s^2\,{}^1S_0$ binding-energy difference. The signature of this "three-photon" decay channel can also be detected by x-ray–x-ray coincidences. Moreover, we emphasize the potentially interesting possibility to utilize RTE to populate the $1s2s\ ^1S_0$ state for studies of its $2E1$ decay.

In the present experiment,[10] beams of hydrogenic $_{32}Ge^{31+}$ ions with specific energies between 12 and 19 MeV/u from the UNILAC facility at GSI Darmstadt impinged after a narrow collimation on a molecular H_2 gas target. The target was an open, three-stage, differentially pumped gas cell, designed in such a way that even at the highest employed pressures of about 1 mbar—monitored by a capacitance manometer—only a small increase of the residual vacuum in the beam line ($\sim 10^{-6}$ mbar) was observed. The reaction volume of the gas target was viewed by two x-ray detectors mounted *vis-à-vis,* perpendicular to the beam direction, outside the gas target which was closed by 6-μm Mylar x-ray windows. The two Si(Li) detectors with 25-μm Be windows had an effective distance of 12 mm from the beam center, viewing an intersection length of the beam with the gas target of 7.5 mm each with solid angles of 1% of 4π. Since the ion velocity was between $0.15c$ and $0.20c$, a time window of about 70 ps could be observed by the x-ray detectors, which has to be compared with a lifetime of 56 ps for the $1s2s\ ^1S_0$ state in Ge^{30+}.[9] The electronics was a standard fast-slow coincidence setup using a time-to-pulse-height converter to measure the time correlation between the fast x-ray signals from both detectors. The ion beam was stopped in a Faraday cup and integrated for normalization. Additionally, Rutherford-scattered particles from a thin Au foil placed in front of the Faraday cup were independently used for beam normalization. Single-collision conditions were ensured by testing the linearity of the reaction rates with gas pressure.

In Fig. 1 a two-dimensional cluster plot of coincident x-ray–x-ray events is displayed for a beam energy of 13.0 MeV/u. The event plot shows a strong "line" with the x-ray energies around $E_1 = E_2 = 10.7$ keV. Additionally, "continua" along three ridges are observed (see the dashed lines). Two of them are parallel to one of the axes and pointing roughly towards the "line"; the third is diagonal, with the sum of both x-ray energies constant, $E_1 + E_2 = \text{const}$ (~ 10.3 keV). At the impact energy of 13 MeV/u we are at the maximum for the *KLL* RTE resonances for the Ge^{31+} projectile. As in this case both of the electrons of the formed intermediate state are in the *L* shell, the first stabilizing radiative transition corresponds to a hypersatellite, $K\alpha^H$ ($L \rightarrow K$ transition in the presence of a further *K* vacancy). The second $L \rightarrow K$ transition is a satellite transition $K\alpha^S$. The mean transition energies calculated for $K\alpha^H$ and $K\alpha^S$ in Ge^{30+} using the multiconfiguration Dirac-Fock program from Ref. 11 are indicated by arrows in Fig. 1. For *KLL* RTE we expect coincident $K\alpha^H$-$K\alpha^S$ (and $K\alpha^S$-$K\alpha^H$) events. Because of Doppler broadening these two narrow-lying peaks melt into the one intense cluster shown in Fig. 1.

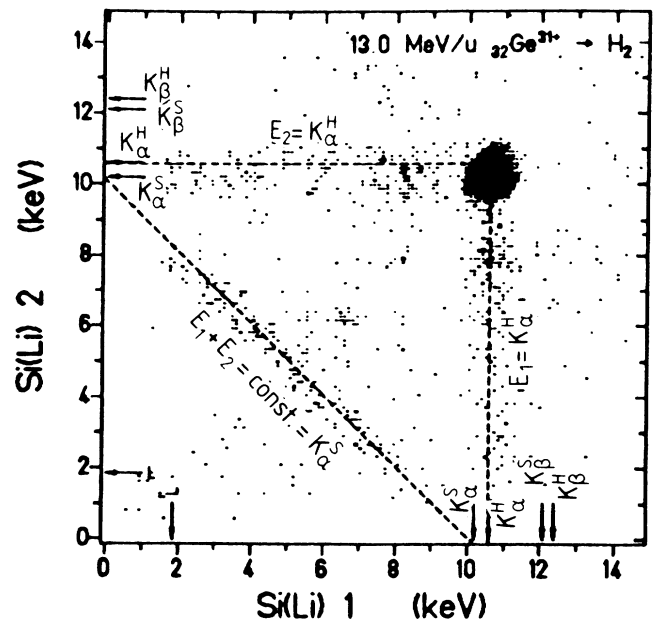

FIG. 1. Two-dimensional plot of the true x-ray–x-ray coincidences at 13.0 MeV/u collision energy. The $K\alpha$-$K\alpha$ coincidences are clearly visible in this event plot. The continuous ridges coming from the two-photon decay ($2E1$) of the metastable $1s2s\ ^1S_0$ state are marked with dashed lines.

We point out that at 13 MeV/u we do not observe any contributions from radiative transitions involving higher shells, e.g., $K\beta^S$, $K\beta^H$, $L\alpha$, etc: As we are at the *KLL* RTE resonance, the absence of particularly the $K\alpha$-$K\beta$ and $K\beta$-$K\beta$ coincidences can be taken as strong evidence for negligible contributions of other nonresonant capture processes leading to the emission of two *K* x rays, e.g., nonresonant transfer and excitation[12] (NTE) and radiative electron capture into higher shells (*n*-REC).[13] They will not populate the *L* shell alone.

The three ridges seen at the *KLL* resonance energy of 13 MeV/u (Fig. 1) cover at least in one detector an energy range from 0 to the full $K\alpha^S$ energy. They are caused by the two-photon decay of the metastable $1s2s\ ^1S_0$ state, which itself is populated by a hypersatellite transition from the doubly excited resonance states. Only two of the three x rays, $K\alpha^H$, E_1, and E_2 (with $E_1 + E_2 = K\alpha^S$ and $E_1, E_2 \leq K\alpha^S$), are detected and lead to the observed ridges $K\alpha^H$-E_2, E_1-$K\alpha^H$, and E_1-E_2. The three continua, i.e., the two-photon decay of the $1s2s\ ^1S_0$ state, are also found for the higher RTE resonance regions (like *KLM*) due to cascading transitions to the $1s2s\ ^1S_0$ level. From the continua one obtains, on the one hand, the spectral distribution for the two-photon decay, giving information on the structure of the two-electron system; on the other hand, the cross section of the involved RTE resonances can be extracted from the total intensity in the ridges. As the ridges parallel to the axis are slightly disturbed by the detector responses from the $K\alpha$ x rays and by some background contributions, we will only use in the following the information contained in the diagonal ridge.

The energy spectrum of one of the two photons obtained from the diagonal ridge is shown in Fig. 2. Because of the uncertainties in coincidence-detection efficiency the spectrum is cut off on both sides, i.e., below 2 and beyond 8 keV; in between, the spectrum is efficiency corrected. The spectrum should be symmetric around $\frac{1}{2}(E_1+E_2)=\frac{1}{2}K\alpha^S$ which is fulfilled within the available low statistics. We mention that a pure excitation of the hydrogenic Ge^{31+} projectile to the $2s\,^2S_{1/2}$ level may also result in a two-photon decay contributing to the diagonal ridge alone. However, several independent checks showed that this contribution is negligible. In Fig. 2 we also plot the energy spectrum expected for the two-photon decay for hydrogen $(Z=1)$.[9] According to calculations of Drake[14] for He-like systems, electron correlation and relativistic effects roughly compensate for our Z region and the H distribution can be used as a simple test. However, in order to get a reliable comparison with theory, dedicated experiments with better statistics at possibly higher Z are needed. The shape of the two-photon decay spectrum contains indeed very detailed information on the structure and dynamics of two-electron systems, i.e., the $1s2s\,^1S_0$ state and its decay. Normally, only the integral over the $2E1$ spectrum, that means only the total decay rate, is measured in order to test theories; see, e.g., Ref. 15.

For completeness we insert in Fig. 2 a decay diagram for the relevant levels in He-like ${}_{32}Ge^{30+}$ restricted to levels accessible for *KLL* RTE processes and decaying via the $1s2s\,^1S_0$ level. For a more complete decay diagram relevant for all *KLL* processes, see Refs. 5 and 16. The numbers given in the inset are calculated using the multiconfiguration Dirac-Fock program from Ref. 11 and an additional subroutine for the radiative dipole transition rates.[17] Because of the intermediate coupling active for Ge there are also intercombination lines. Considering additionally the Auger rates[18] relevant for the production of the intermediate states, one finds that for the *KLL* region the resonance leading to the doubly excited $2s2p\,^1P_1$ state is 98% and the dominant one populating the $1s2s\,^1S_0$ level. Correspondingly, we deduced the decay branches for *KLM* RTE. There, about nine resonances contribute to populate the $1s2s\,^1S_0$ level, with the $2p3p\,^1D_2$ resonance is dominating at 50%.[10]

The $2E1$ transition rate for the $1s2s\,^1S_0$ state given in Fig. 2 is calculated according to Ref. 9. Within the accessible time window of our experimental arrangement the $1s2s\,^1S_0$ state decays with a probability of about 70%. On the other hand, the angular correlation expected for the $2E1$ decay[9] enhances the detection probability by about 30%.

Integrating the intensity in the two-photon spectrum along the diagonal ridge yields the total cross section for the two-photon emission. In Fig. 3 these cross sections for ${}_{32}Ge^{31+}+H_2$ collisions are plotted as a function of projectile energy. The cross sections given are corrected for x-ray detection efficiency, experimental time window, and the angular correlation of the $2E1$ decay. (No further anisotropy in the x-ray emission has been assumed, which seems to be reasonable for our case; cf. Ref. 19.) The error bars give the statistical uncertainties. Additionally, we have to place a normalization uncertainty of about 25%. In the figure the resonance positions are indicated by arrows. The *KLL* resonance is clearly separated from higher-shell resonances *KLn* with $n\geq 3$, having a deep minimum in between. The small cross-

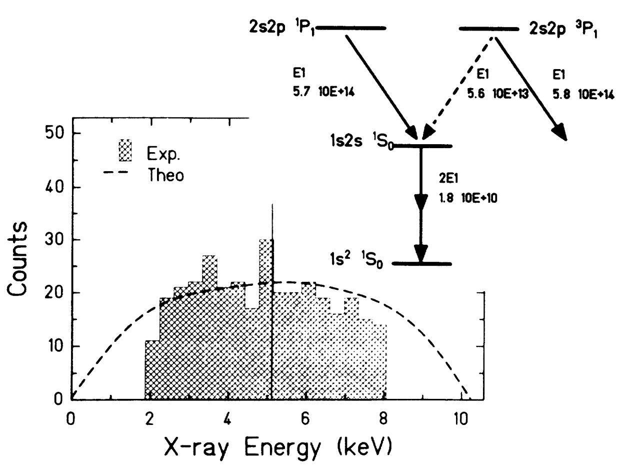

FIG. 2. Energy distribution of one of the two photons from the $2E1$ decay measured in coincidence with each other. The expectation is displayed as the dashed curve; see text. Inset: The level scheme of the He-like ${}_{32}Ge^{30+}$. States which are accessible via *KLL* RTE processes and may decay via the $1s2s\,^1S_0$ state are marked in the *L-S* coupling scheme. Radiative transition rates are given in s^{-1}.

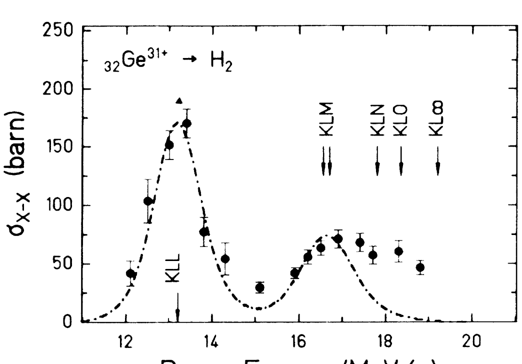

FIG. 3. Excitation function for the ${}_{32}Ge^{31+}+H_2$ collision system. The cross sections for the $2E1$ photon emission (solid circles) are represented as a function of collision energy. The dash-dotted line gives our calculations. The triangle gives an estimate from Ref. 21.

section values in the minimum indicate that competing nonresonant processes possibly leading to a $2E1$ decay are of vanishing importance.

As there are no predictions available in the literature for RTE in $Ge^{31+}+H_2$ collisions we calculated the corresponding cross sections for the *KLL* and *KLM* regions. Using our calculated level and decay diagrams (according to Refs. 11 and 17, respectively) and the tabulated Auger rates from Ref. 18 we determined the DR rates. Folding the corresponding DR rates with the Compton profile for molecular-hydrogen target electrons[20] we obtained the cross sections plotted as a dash-dotted curve in Fig. 3. There is an excellent accordance between experiment and calculation for the *KLL* resonance. An independent calculation for the *KLL* maximum by Hahn and McLaughlin[21] yields about 190 b. This value is represented by the triangle in Fig. 3 and is also in excellent agreement. For the *KLM* region there is also good accordance between our calculations and measurements. For higher-shell resonances there are no calculations available to us at the moment.

In conclusion, using the x-ray–x-ray coincidence technique we isolated a single resonance, the resonance to the $2s2p\,^1P_1$ state in He-like Ge, for resonant transfer and excitation in collisions of H-like Ge with hydrogen. The measured cross sections for this RTE resonance are in excellent agreement with calculations within the impulse approximation; i.e., taking into account the Compton profile for molecular hydrogen, dielectronic recombination and RTE are directly equivalent processes. The $2s2p\,^1P_1$ resonance decays to the metastable $1s2s\,^1S_0$ state and is the only dominant *KLL* resonance feeding this level, which can only decay by two-photon emission to the ground state. It is demonstrated in this Letter that the $2E1$ decay in heavy He-like ions can be effectively studied by means of the x-ray–x-ray coincidence technique and that RTE is the dominant production mechanism populating the $1s2s\,^1S_0$ state. Dedicated experiments to measure the shape of the two-photon decay spectrum will yield detailed information on relativistic, correlation, and QED effects in very heavy two-electron ions.

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