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Direct Observation of $K\alpha$ Hypersatellites in Heavy-Ion Collisions*

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In this paper it is demonstrated that 30-MeV oxygen ions bombarded on Ca produces a $K\alpha$ spectrum consisting of hypersatellite lines. $K\alpha$ hypersatellites are $1s \rightarrow 2p$ transitions in an atom with two $1s$ electron vacancies in the initial state.

We have observed for the first time double K-shell ionization produced by scattering high-energy heavy ions from a target. The production of these ionization states in the target atom is manifested in the observation of $K\alpha$ x-ray transitions ($1s \rightarrow 2p$ transitions) which occur at a higher energy than the normal $K\alpha_{1,2}$ x ray. These transitions have been called hypersatellites and have been observed in K–x-ray, K–x-ray coincidence experiments following either electron capture or internal conversion in a readioactive sample.[1] In the present experiment the $K\alpha$ hypersatellites of Ca are observed in the bombardment of 30-MeV oxygen on Ca and are detected directly in a noncoincidence high-resolution spectrum obtained with a crystal spectrometer. This method allows one to determine very accurately the position of the hypersatellites and the relative intensities of the various transitions.

The experimental situation is quite simple. A 30-MeV O^{5+} beam from the University of Texas tandem accelerator is focused onto the target placed at 45° to the beam. The vacuum crystal spectrometer is placed at 90° to the beam. The data accumulation system is automated and controlled by a PDP-7 computer. Once the spectrometer is positioned at the initial setting, a computer command begins the scan by accumulating counts from the flow proportional counter and from the beam integrator. After a preset beam integration the spectrometer is stepped to its next position and continues the process a preset number of times.

The observed Ca K–x-ray spectrum produced by 30-MeV oxygen is presented in Fig. 1. The peaks labeled A through F are the $K\alpha$ transitions corresponding to initial states $(1s)^{-1}(2p)^{-n}$ for $n=0$, 1, 2, 3, 4, and 5, respectively. The peak A is thus the $K\alpha_{1,2}$ transition and the peaks B through F are the so-called $K\alpha$ satellites. The probability for single-K, multiple-L-shell ionization is therefore seen to be much greater than the probability for single K-shell ionization. This effect was first observed as an energy shift in poor-resolution experiments with Si(Li) solid-state detectors.[2] More recent experiments[3-5] performed with crystal spectrometers have revealed the enhanced K satellite structure as observed in Fig. 1.

The newly observed transitions reported in this experiment are the ones labeled D', E', and F' in Fig. 1 and are the ones which occur above the $K\alpha_{1,2}$ transition at 3916, 3941, and 3968 eV, respectively. These transitions are thus 224, 248, and 274 eV above the $K\alpha_{1,2}$ transition, and 98, 74, and 48 eV below the $K\beta_{1,3}$ transition. Assuming that these transitions are from Ca, they must be $K\alpha$ ($1s \rightarrow 2p$) type transitions and not $K\beta$ ($1s \rightarrow 3p$) type transitions. Higher-energy $K\alpha$ transitions can be created by double K-shell ionization. Hartree-Fock-Slater calculations have been performed for $K\alpha$ transitions in atoms

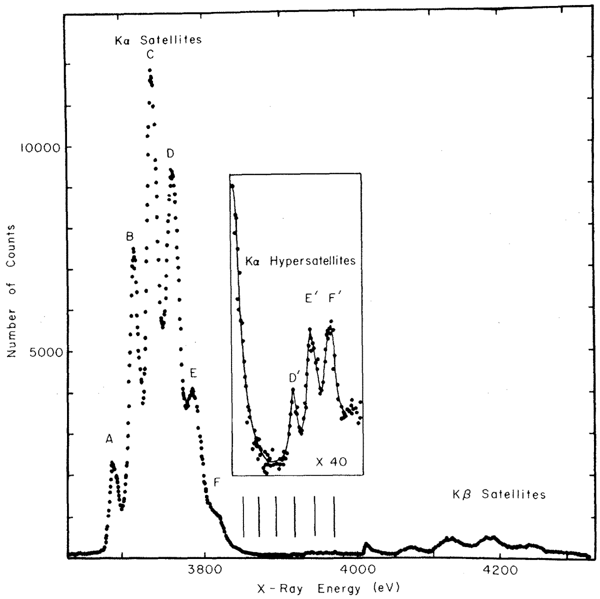

FIG. 1. K–x-ray spectrum of Ca produced by 30-MeV oxygen bombardment. The $K\alpha_{1,2}$ peak is the peak labeled A, whereas peaks B through F are enhanced satellite lines due to single-K, multiple-L-shell ionization. The $K\beta$ spectrum shows a similar structure. The $K\alpha$ hypersatellites due to double K-shell ionization are contained in the inset, in which the counts have been multiplied by a factor of 40. The six vertical lines from left to right show the hfs calculated energies of the $K\alpha$ transitions from initial states $(1s)^{-2}(2p)^{-n}$ for $n = 0$, 1, 2, 3, 4, and 5, respectively.

with initial states $(1s)^{-1}(2p)^{-n}$ and $(1s)^{-2}(2p)^{-n}$ for $n = 0$, 1, 2, 3, 4, and 5. The peaks D', E', and F' agree quite well with the calculated energies for $(1s)^{-2}(2p)^{-3}$, $(1s)^{-2}(2p)^{-4}$, and $(1s)^{-2}(2p)^{-5}$ initial configurations, respectively. On this basis the new lines are designated as the Ca $K\alpha$ hypersatellites.

Table I summarizes the experimental and calculated $K\alpha$-transition energies for Ca. As discussed above, the hypersatellites are tentatively assigned as due to transitions from $(1s)^{-2}(2p)^{-n}$ configurations for $n = 3$, 4, and 5, and no evidence is found for the first three hypersatellites. By comparing the calculated and experimental energies for the $K\alpha$ satellites it is found that the experimental values are ~7 eV higher, which can be due to M-shell vacancies. This therefore suggests that the $K\alpha$ hypersatellites should also occur at energies slightly above the hfs calculated values. More calculations are necessary to further explore this point.

The hypersatellites observed in K capture or internal conversion[1] are observed in poor resolution so that no designation of initial state can be made, but it is presumed that the $(1s)^{-2}$ state is the only one excited. If by hypersatellite one refers specifically to this transition, then the states seen in the present experiment should be properly designated the satellites of the hypersatellite line.

The intensity ratio of $K\alpha$ satellites to $K\alpha$ hypersatellites is measured to be $(K\alpha)^S/(K\alpha)^H = 210 \pm 80$. The quantity $(K\alpha)^S$ includes the $K\alpha_{1,2}$ intensity. In obtaining this ratio no attempt was made to correct for changes in crystal reflectivity with angle. This measured ratio cannot be taken as the ratio of single K to double K ionization since the competing K Auger rates may be slightly different in the two cases.

TABLE I. Observed and calculated energies of $K\alpha$ transitions in Ca.

Configuration of initial state	Energy (eV) Observed	Energy (eV) hfs	$E(K\alpha_{1,2}) - E_{\text{calib}}$ (eV) Observed	$E(K\alpha_{1,2}) - E_{\text{calib}}$ (eV) hfs	
$(1s)^{-1}$	3690[a]	3690	···	···	$K\alpha_{1,2}$
$(1s)^{-1}(2p)^{-1}$	3716	3710	26	20	$K\alpha$ satellites
$(1s)^{-1}(2p)^{-2}$	3738	3732	48	42	
$(1s)^{-1}(2p)^{-3}$	3763	3754	73	64	
$(1s)^{-1}(2p)^{-4}$	3786	3779	96	89	
$(1s)^{-1}(2p)^{-5}$	3814	3806	124	116	
$(1s)^{-2}$	···	3848	···	158	$K\alpha$ hypersatellites
$(1s)^{-2}(2p)^{-1}$	···	3869	···	179	
$(1s)^{-2}(2p)^{-2}$	···	3892	···	202	
$(1s)^{-2}(2p)^{-3}$	3914	3916	224	226	
$(1s)^{-2}(2p)^{-4}$	3938	3941	248	251	
$(1s)^{-2}(2p)^{-5}$	3964	3968	274	278	

[a]Calibration energy taken from J. A. Bearden, Rev. Mod. Phys. 39, 78 (1967).

In order to corroborate the results reported here, similar experiments have been performed on Sc and Ti. In both cases structure very similar to the one reported in this paper was observed. This essentially eliminates the possibility of the peaks being due to target impurities. These results will be reported later in a full length paper. A $K\alpha$ spectrum of Sc was taken under proton bombardment, in which case no structure was seen in the region of the $K\alpha$ hypersatellites. In addition to eliminating the possibility of target impurities, this result eliminates the possibility of the x-ray lines being due to crystal defects and/or geometrical factors.

In summary it has been shown that heavy-ion collisions can be used to excite $K\alpha$ hypersatellites. Three $K\alpha$ hypersatellites of Ca are observed when bombarded with 30-MeV oxygen. The energies of the lines are shown to be consistent with hfs calculated energies of states with double K-shell ionization. The hypersatellite lines are observed to be 0.5% of the total $K\alpha$ spectrum. This observation further demonstrates that heavy ions can be used to give a more complete $K\alpha$ spectrum.

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Transient-Nutation Effects in Time-Resolved Infrared-Microwave Double Resonance of Ammonia*

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Transient nutation is observed in the time-resolved infrared-microwave double resonance of ammonia. This behavior is described by a Bloch equation for relaxation of the ammonia inversion levels, with an effective T_2 cross section of 400 ± 50 Å^2. A similar effect was produced by rapidly switching the Stark field applied to the molecules, which suddenly brings them into resonance with the cw microwave radiation.

The continuing development of coherent, tunable radiation sources, such as microwave klystrons and optical lasers, has opened the possibility of using many of the techniques of magnetic resonance in the study of optical systems. In particular, the method of double resonance, commonly employed in radio-frequency spectroscopy, has proven extremely useful in determining relaxation parameters, and, to a lesser extent, spectrum assignments, in coherently excited rotational and vibrational levels of molecules. Microwave-microwave double resonance[1-3] has been carried out in a large number of systems, and infrared-microwave double resonance in ammonia,[4] methyl chloride,[5] and formaldehyde.[6] Steady-state experiments, such as the aforementioned, are very useful in elucidating the propensity rules for collisional transfer of rotational energy. Time-resolved experiments, on the other hand, are required in order to obtain absolute magnitudes of energy transfer rate constants and transition probabilities. Several time-resolved microwave-microwave[2,3,7] and infrared-microwave[5,8] experiments have been reported, and extensive time-resolved infrared-infrared double resonance has been carried out in SF_6[9] and BCl_3.[10] The rate-constant information derived from such experiments is necessary to assess the range of the interaction potentials postulated to interpret the propensity rules derived from the steady-state experiments.

In this Letter we report the results of a time-resolved infrared-microwave double-resonance study in gaseous ammonia. We observe a transient nutation of the inversion levels of the molecule following the applied infrared pulse. In order to understand this phenomenon, we had to consider the phenomenological Bloch equations,