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Observation of Z = 70 Quasiatomic K X Rays from 30- and 60-MeV  $_{35}Br + _{35}Br$  Collisions\*

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A K-x-ray band up to the united-atom limit for Z = 70 has been observed in 30- and 60-MeV Br+Br collisions. This band can be assigned unambiguously to transitions between molecular orbitals. With a KBr target, yields at the two energies are (for  $E_x \ge 22$  keV)  $2 \times 10^{-8}$  and  $10^{-7}$  photons/projectile, respectively, or  $10^{-4}$  photons/beam K vacancy, nearly independent of beam energy. Much reduced yields are found for Ti, Fe, and Zr targets.

Thick, solid KBr targets were bombarded with 30- and 60-MeV Br. A quasiatomic x-ray band extending up to the united-atom<sup>1</sup> K-x-ray limit for Z = 70 was observed. To identify the band unambiguously, we have demonstrated that (1) it is not due to electronic pileup, (2) it is not due to a heavy impurity, (3) it is considerably reduced in intensity if the target has a few units higher or lower Z than the beam, (4) it has the expected high-energy cutoff (52-59 keV), and (5) it has the expected total intensity. Previously observed x-ray bands have been assigned to quasiatomic M,<sup>2</sup> L,<sup>3</sup> and K x rays<sup>4</sup> without such detailed identification.

In our arrangement a  $17-cm^3$  Ge(Li) detector viewed the target surface through a 1.6-mm Lucite window in front of which Al absorbers could be placed. Since for these experiments it was possible (as shown below) to run at sufficiently low counting rates such that pulse pileup could be neglected, no electronic pileup rejection was used. This avoided any possibility of spectrum

distortion from such rejection schemes at these low x-ray energies. Figure 1 shows three spectra each for a KBr and a Zr target. With no absorber (spectrum a) a pileup "peak" is seen at approximately double the energy of the main Br K-x-ray (12 keV) peak. Insertion of 0.8-mm Al absorber (spectrum b) reduced the Br K-x-ray peak approximately by the expected factor  $\frac{1}{40}$ ; hence, the pileup peak should have been reduced by  $\frac{1}{1600}$ , i.e., to negligible proportions. To demonstrate that this was indeed accomplished, a further 0.8-mm Al absorber was inserted (spectrum c). Now the spectrum above the main K-xray peak was reduced only by the amount appropriate to the particular x-ray energy (see curve d). All spectra shown in subsequent figures have been taken with 1.6-mm Al absorber, and hence do not contain pileup effects (counting rates were below 50/sec).

In Fig. 2 linear plots of the uncorrected spectra are shown. Vertical lines mark the expected end points of the quasiatomic  $K_{\alpha}$  and  $K_{\beta}$  x rays.



FIG. 1. K-x-ray spectra from KBr and Zr targets bombarded with 30-MeV Br beam. Spectrum a, no absorber, spectrum b, 0.8-mm Al absorber, spectrum c, 1.6-mm Al absorber, curve d, calculated absorption factor for 0.8-mm Al with abscissa representing unity (see left scale). Arrows indicate expected pileup peaks, vertical lines show calculated  $K\alpha$  and  $K\beta$  energies for Br, Zr, Z = 70, and Z = 75.

Only room background has been subtracted (except for the 60-MeV spectra—see below). We have not yet been able to investigate carefully whether any of the spectra contain additional, beam-induced background; even if they do, the difference between KBr and the other targets is obvious, and is in qualitative agreement with expected intensity ratios, as discussed below.

The KBr targets were deposited on 0.025-mm Al foils. Although some flaking off occurred during bombardment, three different targets gave similar results. At 60 MeV, though, a clear beam-induced background was found with a pure Al-foil target. This background has been subtracted in Fig. 2(c) (crosses). It is satisfying that the 30- and 60-MeV Br+KBr spectra have similar shapes and merge into background at the same, expected energy. [In both cases the distance of closest approach between the Br nuclei (120 and 60 F, respectively) lies well within the



FIG. 2. Quasiatomic K-x-ray spectra for various targets, with 1.6-mm Al absorber, for 100- $\mu$ C accumulated charge. Solid circles, 30-MeV Br (charge 5+); crosses, 60-MeV Br (charge 7+) (right ordinate scale). Arrows give  $K\alpha$  and  $K\beta$  end-point energies for quasiatomic K x rays.

1s Bohr orbit of the united atom (760 F).]

The insert in Fig. 2(c) shows part of the spectrum from a Ti + Ta target bombarded with 60-MeV Br. The Ta K x rays ( $\alpha$ , 57.5;  $\beta$ , 63.2 keV) appear as *line* spectra. Therefore, the Br + KBr continuum spectra cannot be due to heavy impurities. Also, one sees from a comparison of Figs. 2(a) and 2(c) that the potassium, which has a Z close to that of Ti, should contribute only little to the KBr spectra.

After correction for absorption and detectionefficiency effects, the Br + KBr spectra shown in Fig. 3 are obtained. The approximately exponential falloff of the spectra is similar to that found for C+C collisions.<sup>4</sup> Table I summarizes the total quasiatomic x-ray yields ( $E_x \ge 22$  keV) found. The Br + KBr yield relative to the Br-beam Kvacancy yield is approximately 10<sup>-4</sup>, independent of beam energy. (For the other targets the yields given must be considered as upper limits in view of possible beam-induced background.)

In an energetic, close collision between two atoms, with Z not too different, the K level of the higher-Z collision partner becomes the  $1s_{1/2}$ ,



FIG. 3. Absolute quasiatomic K-x-ray spectra for Br+KBr, corrected for absorption and detection-efficiency effects. Arrows indicate quasiatomic  $K\alpha$  and  $K\beta$  x-ray end points for Z = 70. Present statistical errors do not allow any particular significance to be attached to the humps in the spectra above 50 keV.

and the K level of the lower-Z partner, the  $2p_{1/2}$ level of the united atom.<sup>5</sup> This has two consequences. (1) Observation of a guasiatomic (united atom or nearly united atom)  $K \ge ray$  requires a K vacancy in the *higher-Z* partner either before or during the collision. (2) Electron promotion, which can be important in generating a vacancy in the  $2p_{1/2}$  molecular orbital (MO) if the colliding Z's are similar, is ineffective in producing quasiatomic K x rays, except in symmetrical or nearly symmetrical collisions, but then only by a two-step process. In the latter case, a K vacancy created by electron promotion in a first collision may be carried into a second collision where it may produce a quasiatomic  $K \ge ray$ . This is the mechanism described in Refs. 2 and 3; its operation requires the use of solid targets because typically K vacancies are carried by the projectile only over a distance of the order of 10 atomic diameters. Since the probability of the second collision coming within a K orbit is very small (see below), in addition to the two-step process we must also consider a onestep process in which the  $1s_{1/2}$  MO is (Coulomb) excited and, during the same collision, decays with emission of a quasiatomic  $K \ge ray$ .

Lacking a detailed theory,<sup>6</sup> we present a very crude, order-of-magnitude estimate of the ex-

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E (MeV)	Target	Relative yield <sup>a</sup>	Absolute yield <sup>b</sup>
30	KBr <sup>c</sup>	90 <sup>d</sup>	17
60	KBr <sup>e</sup>	100 <sup>d</sup>	110
30	Ti <sup>c</sup>	≤ 2000	≤4
30	Fe <sup>ċ</sup>	≤ 1000	≤ 9
30	Zr <sup>c</sup>	≤ 5 <sup>f</sup>	≤ 5

<sup>a</sup>Yield for  $E_x \ge 22$  keV per 10<sup>6</sup> beam K vacancies (maximum error = ± 50%). Assumes Br fluorescence yield = 0.62.

<sup>b</sup>Yield for  $E_x \ge 22$  keV per 10<sup>9</sup> projectiles (maximum error = ± 50%).

<sup>c</sup>Room background only subtracted.

- <sup>d</sup>Assumes beam K vacancies  $=\frac{1}{2}$  total Br K vacancies.
- <sup>e</sup>Subtraction made for background from Al backing.

<sup>f</sup> Yield per  $10^6$  higher-Z partner (Zr) K vacancies is ~ 70 times larger (see Table II).

pected quasiatomic K-x-ray yields by the two processes. Considering first a symmetric collision, let  $Y_s$  and  $Y_p$  be the  $1s_{1/2}$  and  $2p_{1/2}$  MO vacancy yield per projectile. Then the total K-vacancy yield of the isolated projectile is

$$Y_K = Y_s + Y_p \,. \tag{1}$$

The total quasiatomic *K*-vacancy yield is, roughly,

$$Y_{Q} \approx Y_{s} t / \tau' + Y_{K} (v \tau / d) P_{K} t / \tau', \qquad (2)$$

where the first term on the right-hand side represents the one-step process with emission of a quasiatomic K x ray (or Auger electron) during the collision time t. The mean life of a K vacancy is denoted by  $\tau$  and  $\tau'$  for separated and "quasi" atoms, respectively. The second term on the right-hand side represents the two-step process, with v the speed of the projectile, d is the distance between atomic planes, and  $P_K$  is the probability of overlap of projectile and target K orbits in one atomic plane.

For quasiatomic K-vacancy formation in asymmetric collisions,  $Y_s$  and  $Y_p$  in Eq. (2) must refer to the higher-Z partner since the vacancy must occur there. As the difference between the Z's of the collision partners increases,  $Y_p/Y_s$  drops rapidly to zero<sup>7,8</sup> and the first term of Eq. (2) will be dominant because  $(v\tau/d)P_K \ll 1$  (see below).

We now apply Eq. (2) to 30-MeV Br + Br collisions. From a study of 47-MeV I+I collisions<sup>8</sup> we infer that  $Y_s/Y_K \simeq 10^{-3}$  to  $10^{-2}$ . We assume the same relationship holds in the present case

TABLE II.	Relative K-x-ray	yields	for	higher-Z	col-
lision partner	in Br collisions.				

Target	Relative yield <sup>a</sup>
22 <b>Ti</b> 26Fe 35 <b>Br</b> 40 <b>Z</b> r	$     1 \times 10^{-3}      7 \times 10^{-3}, 2.5 \times 10^{-2^{b}}      1      4 \times 10^{-2} $

<sup>a</sup>From 45-MeV Br cross-section measurements of Ref. 10, except as noted (error  $= \pm 30\%$  maximum).

<sup>b</sup>From 30-MeV Br yield, determined by us (error = ± 50% maximum).

and that  $t \simeq 4 \times 10^{-19}$  sec,  $\tau \simeq 3 \times 10^{-16}$  sec,<sup>9</sup> and  $\tau' \simeq 4 \times 10^{-17}$  sec for a mean quasiatomic x-ray energy of 28 keV, obtained from Fig. 3. From the geometry of the KBr crystal structure we find  $(v\tau/d) \simeq 10$  and  $P_K \simeq 10^{-4}$ . Hence,

$$Y_Q \simeq [(10^{-5} \text{ to } 10^{-4}) + 10^{-5}] Y_K.$$
 (3)

The first term in the bracket refers to the onestep process, the second to the two-step process. Our crude estimates cannot decide the relative importance of the two processes. In order of magnitude, the ratio  $Y_Q/Y_K$  agrees with the experimental value of ~10<sup>-4</sup> (Table I).

For the asymmetric collisions, two main effects occur: (1) Since  $Y_K \rightarrow Y_s$ , the one-step process should dominate and  $Y_Q/Y_K$  should increase to ~10<sup>-2</sup>. (2) Since  $Y_K$  for the higher-Z partner decreases extremely rapidly in magnitude<sup>10</sup> as the Z's differ (see Table II),  $Y_Q$  should drop rapidly in magnitude. These two predictions are indeed approached experimentally (Table I).

The method outlined here for the unambiguous identification of quasiatomic K x rays can be extended to heavier ions. In particular, in searching for the (atomic) internal pair-conversion process predicted by Müller and co-workers<sup>11</sup> for  $Z_1 + Z_2 \ge 169$ , one can use a U beam and bombard,

in succession, Th, U, and Pu targets. The maximum yield should occur with a U target and much reduced yields with Pu and Th.

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