investigated in a forthcoming paper. Since these interference effects show up where the phase difference between contributions from approaching and leaving ions along the scattering trajectory are largest, they should occur most pronounced for low-energy transitions and their oscillation frequency should increase with bombarding energy of the ions.

[10]We acknowledge help in the calculations by W. Betz.

Evidence for Quasimolecular K X-Ray Emission in Heavy-Ion Collisions from the Observation of the X-Ray Directional Anisotropy*

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We have observed an anisotropic angular distribution for the high-energy continuum x rays emitted in high-energy collisions of Ni ions with Ni atoms. The variation of the anisotropy with photon energy is uniquely characteristic of induced radiative transitions in quasimolecules, and hence provides strong evidence for the quasimolecular origin of these radiations.

A search for the formation of intermediate electronic molecules in close collisions of very heavy ions has been motivated recently by the prospect that such united atom phenomena may ultimately provide access to the spectroscopy of superheavy atoms and, therefore, to fundamental processes associated with the quantum electrodynamics of strong fields.[1] Proof for the existence of such quasimolecules has been sought in the emission of K x-ray radiation from transitions between the molecular orbitals that may be formed in the collision.[2-6] In this paper we report on the results of an investigation to detect such transitions in the ^{58}Ni + ^{58}Ni ion-atom system colliding at high energy. We discuss particularly the observation, for the first time, of a directional asymmetry for the high-energy continuum x rays emitted in the collision, which we utilize as a distinctive characteristic for identifying the quasimolecular origin of these radiations.

Studies with light-ion-atom systems have provided some of the more convincing demonstrations of molecular orbital x-ray emission.[2,5] However, the interpretation of experiments involving high-energy collisions of heavy-ion-atom systems,[3,4] which are more relevant to the future investigations noted above, are presently more speculative, relying on an ability to extrapolate detailed identifying features from mechanisms found applicable previously in light systems. The interpretation of these experiments is particularly complicated by the exponential energy dependence of the continuous spectra observed, by uncertainties with regard to the mechanisms dominating K-vacancy production during high-energy heavy-ion collisions, by collision broadening effects, and most important, by the lack of a definitive signature for distinguishing this radiative process from other competing radiations.[4]

Recently, Müller, Smith, and Greiner[7] have noted that the characteristic angular distribution expected for molecular orbital K x-ray emission furnishes such a unique signature. This suggestion centers on the existence of the important, but previously neglected, induced transitions between molecular orbitals which specifically reflect the nonstationary character of the molecular electronic states as they adjust to the rotation of the internuclear axis. Reflecting the nuclear motion, the induced transitions can only be associated with the formation of quasimolecules. They add incoherently to the spontaneous molecular transitions with a relative intensity that grows with increasing photon energy. In the absence of a spatial alignment of the molecular orbitals, the induced radiations are predicted to exhibit an angular emission pattern relative to the ion-beam axis of the form $\frac{1}{2}+\frac{1}{4}\sin^2\theta$, while the spontaneous radiations are expected to be isotropic.[7] Thus the incoherent sum of the two produces a prominent enhancement of the angular distribution asymmetry near the united atom K x-ray energy.[8] As demonstrated in Ref. 8 we note that although an alignment of the molecular orbitals may modify these features quantitatively by changing the angular distributions of both the induced and spontaneous transitions, the observation of an anisotropy which peaks near the united atom K x-ray energy can only imply the existence of induced transitions, and thus of intermediate molecular states. The presence of an alignment of molecular orbitals, as reflected in the angular distribution, in itself speaks for the existence of intermediate molecules.

We have observed an anisotropic angular distribution for the high-energy continuum x rays produced in ^{58}Ni + ^{58}Ni collisions which favors photon emission at 90° over 0°, measured with respect to the ion-beam axis. The anisotropy also exhibits the enhancement, referred to above, for the higher energy x rays. By eliminating the nucleus-nucleus electric dipole bremsstrahlung with the utilization of monoisotopic targets and projectiles, and by accentuating the high-energy portion of the spectra with an Al filter, we were able to extend the investigation of the x-ray continuum over an intensity range of more than 3 orders of magnitude, revealing important features of the high-energy tail. The selection of identical nuclei for target and projectile not only suppressed the nuclear bremsstrahlung background, but equally important, it ensured the absence of radiations from coherent interference of nuclear dipole and quadrupole bremsstrahlung.[9] Unlike the individual multipoles, which produce an excess of radiation at 0° relative to 90°, the anisotropy from their interference has the same sign as the induced transitions.[9]

Figure 1 illustrates x-ray spectra obtained with a thin target (~200 μg/cm^2) after subtractions of a small (~5 counts/channel) ambient background and still smaller contributions from nuclear Coulomb excitation and nuclear bremsstrahlung. The total calculated bremsstrahlung intensity shown in Fig. 1 includes approximately equal parts of quadrupole radiation from the target and bremsstrahlung from a Be beam stop.[9] It is clear that all these backgrounds produce only a negligible distortion of the spectra. Other processes contributing to the x-ray continuum include (1) electron bremsstrahlung,[10] (2) radiative electron capture,[10,11] and (3) collision broadening of the Ni K_α, K_β lines.[12] Their significant effect on the spectra, however, is confined to photon energies below ~15 keV. The interesting small oscillations in photon intensity, clearly visible in the spectra for energies below 15 keV, probably reflect interference effects from the phase difference between radiation from approaching and outgoing ions along the scattering trajectory[8]; this effect will be discussed in more detail in a forthcoming paper. We have limited the

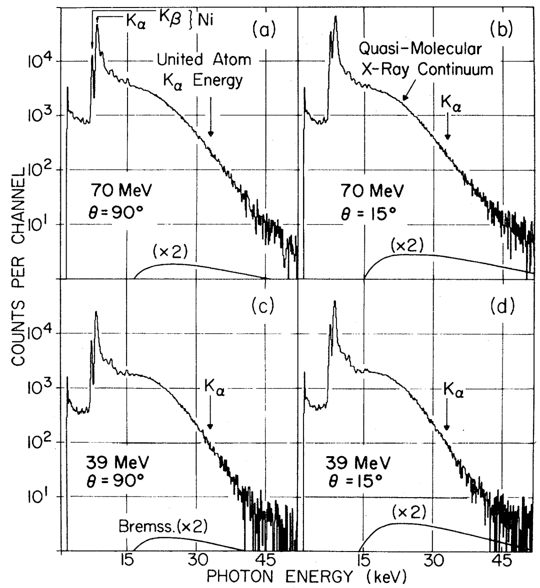

FIG. 1. (a), (b) Unnormalized x-ray spectra taken at bombarding energies 70 MeV and (c), (d) 39 MeV; (a), (c) detector angles are 90° and (b), (d) 15°. Detector resolution is 250 eV at 6.4 keV. The K_α and K_β lines are attenuated by factors of 3.5×10^{-6} and 1.4×10^{-4}, respectively.

present discussion to photon energies above 15 keV to avoid these complicating features and the competing backgrounds mentioned.

Above 15 keV the spectra shown decrease almost exponentially with increasing energy after they are corrected for distortion by the Al filter and counter efficiency. With a range of 3 orders of magnitude in intensity available before background is encountered, it is clear that the high-energy continuum extends beyond the united atom K_α energy of ~32 keV by a considerable amount, and that the extent of the tail depends on the projectile energy. Müller and Greiner[8] have shown that dynamic broadening[12] with an effective collision width of a few keV can completely account for the extensive tails we observe. A collision width of a few keV is reasonable for the collision times encountered in this experiment. It is important to note, therefore, that with sufficient suppression of background, dynamic broadening of the molecular-orbital spectra shifts the apparent end point to a degree determined by the projectile velocity and counting statistics. This is particularly relevant for high-energy collisions where such broadening can obscure the identification of transition frequencies associated with the united atom limit, thus limiting the value of a straightforward utilization of the classical united atom limit for identifying quasimolecular formation.

The angular distribution asymmetries for photon energies above 15 keV were obtained from the measured spectra after they were corrected for absorption, counter efficiency, and Doppler-shift effects. An absolute beam integration was provided by an electron-suppressed Faraday chamber, and the relative normalization was corroborated by another x-ray detector which was fixed in position. We define the asymmetry as the intensity ratio $[I(90°)/I(15°)-1]$, where the angles are measured relative to the beam axis, and the photon interval summed is 5 keV. The results are summarized in Fig. 2 as a function of photon energy for bombarding energies of 39 and 70 MeV.

The Doppler-shift corrections are particularly important because of the exponential dependence of the photon intensity with energy. We must note that the calculation of the relevant Doppler shift involves assumptions regarding the mechanisms that dominate the molecular-orbital x-ray production. In the event that the K-vacancy production and molecular radiative relaxation occur in a single ion-atom encounter, it is clear that the relevant velocity is the velocity of the center of mass. This one-step mechanism, therefore, is associated with the maximum possible Doppler shift. The calculation of the Doppler-shift effect for a two-step process (K-vacancy production in one collision followed by the emission of a molecular orbital x ray in a second encounter of either the scattered projectile or the recoiling atom) involves the impact-parameter dependence of K-vacancy production in the first step. We have extracted an average effective velocity for the latter case, by analyzing the Doppler broadening and shift[13] of the characteristic Ni K x rays which reflect the K-vacancy production processes. This analysis indicates that the mean scattering angle for K-vacancy production is generally small; for example, it is $\lesssim 10°$ for 70-MeV projectiles, which agrees with the trends from other recent studies[14] and with the Massey criterion that the ionization centers around impact parameters $\hbar v/E_K$, where E_K is the K binding energy. Therefore the participation of the scattered projectile in the two-step process produces a Doppler shift which is only marginally smaller than the shift for the one-step process, while the contribution from the much slower laterally recoiling atoms reduces the average shift particularly for the

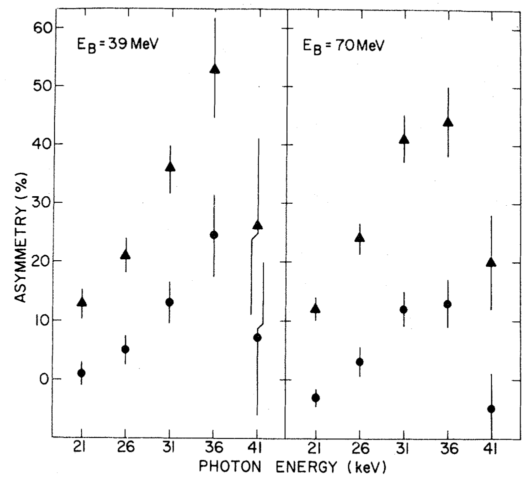

FIG. 2. Asymmetry $\equiv [I(90°)/I(15°)-1]$; triangles and circles are for maximum and no Doppler shift corrections, respectively.

lower energy photons in the spectrum. Figure 2 shows the asymmetries uncorrected for Doppler shift as well as asymmetries which have been corrected for the maximum Doppler shift. It is evident that even the uncorrected asymmetries are large, and that in both extreme cases the asymmetries exhibit the characteristic peaking near the united atom K x-ray energy. Although there is presently some uncertainty as to the exact Doppler-shift correction to be applied at each bombarding energy due to a lack of precise information on the relative importance of one- and two-step processes, it is clear, nevertheless, that the asymmetries lie between the two extreme limits shown in Fig. 2, and that they display the important qualitative features expected for induced quasimolecular transitions.

In addition to the data presented herein, we have also investigated the production of the x-ray continuum for bombarding energies from 5 to 70 MeV. These studies will be the subject of a forthcoming paper. For the present we note that these data display the expected changes in the structure of the high-energy x-ray tail predicted by relativistic calculations with the two-center potential by Müller and Greiner.[8] All these observations reflect the distinctive features that are associated with formation of intermediate molecules, and in particular, the measured asymmetries provide convincing evidence that the continuous spectra observed in these measurements are indeed quasimolecular in origin.

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Anisotropic Emission of Noncharacteristic X Rays from Low-Energy I-Au Collisions

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The x-ray emission for 6- to 60-MeV iodine bombardment on thin Au targets was measured. Spectra, cross sections, and angular dependence of x-ray emission are reported. Special emphasis is placed on the noncharacteristic radiation formerly interpreted as M radiation of a quasiatom with an effective atomic number of 132. The emission of this radiation is found to be nonisotropic strongly supporting the model of induced emission in the collision molecule.

X-ray spectra from heavy-ion–atom collisions may show both characteristic lines of the separated collision partners, as well as broad noncharacteristic bands. In slow collisions the noncharacteristic x rays have been ascribed to radiative transitions between molecular orbitals (MO's) of transiently formed collision molecules.[1,2] Such noncharacteristic x rays, for instance, were observed at 11-MeV iodine bombardment on thick gold targets.[3,4] In these measurements the noncharacteristic band revealed a broad peak at about 8 keV. This energy corresponds to $4f-3d$ transitions[5] in a united atom. Therefore, the x-ray band has been interpreted as M radiation from a short-lived "superheavy quasiatom" with an effective atomic number $Z=Z_1+Z_2=132$. A double-collision mechanism is assumed to be responsible for these x rays. In a first collision an L vacancy is produced in the moving I ion. In a second collision this inner-shell vacancy can decay in the I-Au collision molecule. The existence, as well as the origin, of these x rays has been confirmed by measurements at the University of Rochester.[6] Nevertheless, this MO x-ray emission is still not completely understood. Two critical features remain unexplained: first the high