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in the latter the heavy-ion DWBA accounts for a much smaller fraction of the observed cross section than does the light-ion DWBA. It may be relevant that the ground-state Q value for the reaction  ${}^{48}Ca({}^{16}O, {}^{18}O){}^{46}Ca$  is about 3 MeV more negative than the optimum for momentum matching. In any case, it is clear that anomalies such as this will have to be resolved before two-nucleon transfer data can yield reliable information about the shell-model composition of nuclear wave functions.

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## Periodic Anisotropy Fluctuations in Quasimolecular X-Ray Emission\*

W. Wölfli, Ch. Stoller, G. Bonani, M. Stöckli, and M. Suter

Laboratorium für Kernphysik, Eidgenössische Technische Hochschule, 8049 Zürich, Switzerland

and

## W. Däppen

## Institut für Theoretische Physik, Eidgenössische Technische Hochschule, 8049 Zürich, Switzerland (Received 29 September 1975)

We have found striking periodic intensity fluctuations in the anisotropy of noncharacteristic x-ray radiation emitted in Ca-Ca, Fe-Fe, and Ni-Ni collisions. It is shown that the properties of this new effect can be interpreted as quasimolecular transitions in a rotating two-center system.

It is already known that dynamic effects play an important role in slow heavy-ion-atom collisions<sup>1</sup> and are, e.g., responsible for the large anisotropy of the quasimolecular [molecular orbital (MO)] x-ray radiation observed already by several groups.<sup>2-5</sup> It was also predicted<sup>6</sup> that a sudden rearrangement of the orbital populations in the transient molecular system might produce periodic intensity oscillations superimposed on the continuous MO-x-ray spectra. We report here that such a periodic structure was found during a systematic investigation of the properties of the MO-x-ray spectra produced in Ca-Ca, FeFe, and Ni-Ni collisions at several beam energies. These oscillations, however, are different from those already reported by Smith *et al.*<sup>6</sup> First of all they appear in a different part of the x-ray spectra, namely in the neighborhood of the corresponding united-atom transition energies and not adjacent to the *K* transition energies of the separated atoms. Second, we have already shown in a previous paper<sup>7</sup> that at least at beam energies below 40 MeV only a few weak lines exist in the energy range investigated by Smith *et al.* These lines were proven to belong to an atomic effect and not to a molecular one. The properties of the present new phenomenon are much more visible in the anisotropy than in the singles spectra. Therefore we should like to confine the following discussion to these results.

The experimental arrangement has already been described elsewhere.<sup>7</sup> In brief, momentumanalyzed beams of <sup>40</sup>Ca, <sup>56</sup>Fe, and <sup>58</sup>Ni produced in a Middleton-type ion source and accelerated by the Eidgenössische Technische Hochschule EN tandem accelerator were used to induce the x rays in natural CaF (200  $\mu$ g/cm<sup>2</sup>), isotopically pure  ${}^{56}$ Fe (428  $\mu$ g/cm<sup>2</sup>), and  ${}^{58}$ Ni (440  $\mu$ g/cm<sup>2</sup>) foils. The x-ray spectra were measured with an 80-mm<sup>2</sup>, 5-mm-thick Si(Li) detector (resolution 170 eV at 5.9 keV) at 90° and 30° with respect to the beam axis. An intrinsic Ge x-ray detector  $(80 \text{ mm}^2, 3 \text{ mm thick})$  placed at a fixed angle of  $90^{\circ}$  on the opposite side of the beam axis served as a monitor. The targets were positioned in such a way that self-absorption of the x rays was equal at both angles of detection. The following evaluation procedure was used to determine the anisotropy. First of all, the measured spectra were corrected for the Doppler shift due to the center-of-mass velocity of the colliding system.<sup>8</sup> Then energy windows of variable width were set into the corrected spectra and the total number of counts within each window  $I(E_x, \theta)$  was evalu-



FIG. 1. Global x-ray anisotropies  $I(90^\circ)/I(30^\circ)-1$  for Ca-Ca, Fe-Fe, and Ni-Ni collisions obtained with a window width of 5 keV. In the evaluation, a Doppler shift due to the center-of-mass motion was taken into account. The energies of the  $K\alpha$  transitions in the united atom are indicated by arrows. The position of the anisotropy bump is a function of the total charge of the colliding system. The curves are drawn only to guide the eye.

ated as a function of the x-ray energy. Finally the anisotropy was calculated according to the expression  $I(E_x, 90^\circ)/I(E_x, 30^\circ) - 1$ .

To give first an idea of the general trend of the observed anisotropies, the results of an evaluation with a rather large window width of 5 keV are displayed in Fig. 1. In this representation only a broad anisotropy bump with a badly defined maximum slightly above the united-atom  $K\alpha$  transition energy can be seen. We note that the position and the intensity of these bumps depend on the total charge of the colliding system but practically not on the beam energy in the investigated range between 24 and 40 MeV. This property is in agreement with Ni-Ni results reported recently by Greenberg, Davis, and Vincent<sup>2</sup> and indicates that the originally proposed Coriolis effect is not responsible for the large observed anisotropies.

The most interesting property of the anisotropy, however, can only be seen with a much better energy resolution. The example in Fig. 2 shows distinct periodic intensity fluctuations which appear surprisingly even far beyond the K line of the united atom. Similar fluctuations, not shown here, have been found in the two other cases. To



FIG. 2. Fine structure of the x-ray anisotropy  $I(90^{\circ})/I(30^{\circ})-1$  observed in Fe-Fe collisions at two different beam energies. In both cases the periodic intensity fluctuations are clearly visible if the evaluation is performed with much smaller energy windows.

demonstrate the periodicity of these fluctuations we labeled the minima by integers N starting from the first statistically significant minimum at the low-energy end. If the energies of the minima are plotted against their number N, then all points lie within experimental uncertainty on a straight line. Therefore the energies of the minima can be written as  $E_x^{\ N} = E^0 + N\Delta E$ . The slope  $\Delta E$  of the lines was found to be proportional to  $v_p Z$ , where  $v_p$  is the velocity of the beam (projectiles) and Z is the charge of a single nucleus, whereas  $E^0$  is proportional to  $Z^2$ .

We should like to mention that an examination of the targets with an ultrasensitive proton x-ray analysis did not reveal any disturbing characteristic lines in the energy range of interest. Since no background effect is known which can produce such a periodic structure in the anisotropy, one might be tempted to explain it in terms of the rearrangement effect, proposed first by Smith et al.<sup>6</sup> But in the following, we should like to mention that the observed properties could also reflect the existence of an entirely new molecular effect not considered so far. Our interpretation is based on the fact that any molecular state formed during a heavy-ion-atom collision will split into a set of equidistant substates because of the rotational motion of the guasimolecule. Since this splitting is sufficiently well defined during the collision time, radiative transitions between these states can occur. The energies of these transitions will not be completely smeared out, if the transitions occur preferentially at one selected internuclear distance.

To demonstrate the rotational splitting of the molecular levels, we consider first the simplest possible model of two colliding nuclei each of charge Z and one electron which is forced to follow the motion of the charges. To simplify the problem the collision process is approximated by a uniform rotation of the two nuclei. With units of length scaled by 1/Z and energies by  $Z^2$  the following time-dependent Hamiltonian describes the interaction between the electron and the rotating charges in the nonrotating center-ofmass frame ( $\hbar = 1$ ):

$$H(t) = -(1/2m)\Delta + V(R_{-t}\vec{x}).$$
 (1)

 $V(R_{-t}\vec{x})$  denotes the rotating two-center Coulomb potential.  $R_{-t}$  is the rotation matrix and  $\vec{x}$  the coordinate of the electron in the nonrotating c.m. frame. Note that the present problem is different from that of the ordinary  $H_2^+$  molecule. Since the influence of the electron on the nuclear motion is negligible in our case, the three-body problem reduces to a one-body problem with the given time-dependent Hamiltonian (1).

The time-dependent Schrödinger equation can be solved by the following *Ansatz*:

$$\psi(t, \vec{\mathbf{x}}) = e^{-iEt} u(R_{-t} \vec{\mathbf{x}}).$$
<sup>(2)</sup>

It is shown elsewhere<sup>9</sup> that u satisfies the following eigenvalue problem:

$$[-(1/2m)\Delta + V(\vec{\mathbf{x}}) - (\vec{\Omega} \cdot \vec{\mathbf{x}} \times \nabla)]u(\vec{\mathbf{x}}) = Eu(\vec{\mathbf{x}}).$$
(3)

Since the operator is self-adjoint (although not bounded from below) there exists a complete set of eigenfunctions  $u_v$  belonging to the respective eigenvalues  $E_v$ .

We should like to emphasize that the eigenvalues  $E_v$  of Eqs. (2) and (3) do not have the usual simple physical meaning, because  $u_v(R_{-t}\vec{x})$  is time dependent. Since  $u_v$  is periodic in time, it can be expanded into a Fourier series:

$$u_{\nu}(R_{-t}\vec{\mathbf{x}}) = \sum_{m=0,\pm1,\pm2,\ldots} e^{im\Omega t} u_{\nu}^{m}(\vec{\mathbf{x}}).$$
(4)

Therefore every solution of type (2) can be written as

$$\psi_{\nu}(t,\vec{\mathbf{x}}) = \sum_{m} e^{-i(E-m\,\Omega)\,t} u_{\nu}^{\ m}(\vec{\mathbf{x}}).$$
(5)

This series is the time analog to the Bloch wave solution for a potential periodic in space. There is no limitation to the number of terms, because the two-center potential is not symmetric around the rotation axis. The additional energy term which appears in Eq. (5) indicates that the electron not only gains molecular-orbital energy during a collision, but also a discrete amount of rotational energy equal to  $\Delta E = m\hbar \Omega$ . It is important to note here that this splitting of the molecular states should be observable even if the molecular axis rotates only for a fraction of a revolution. This is due to the fact that the collision time during which molecular levels can be formed (about 10<sup>-18</sup> sec in our cases) is still about one order of magnitude larger than the classical revolution time of an electron in the molecular ground state.

Under the assumption now that the observed periodic structure is the result of radiative transitions into the split molecular ground state, the internuclear distance at which these transitions occur with an enhanced probability can be determined from the experiment (Fig. 3). The energy difference  $\Delta E$  between two substates must be equal to the distance between two subsequent anisotropy minima (or maxima). Expressing this



FIG. 3. Plot of the energies of the minima versus their respective numbers observed in the three cases. The slope of the straight lines clearly depends on the beam energy. The uncertainty of the energy determination of the minima is smaller than the dots used in the figure.

distance in units of the Bohr radius,  $a = \gamma a_0 Z^{-1}$  $(a_0$  is the Bohr radius of hydrogen), we find that all six cases investigated so far (Ca-Ca, Fe-Fe, and Ni-Ni each at two different energies) give almost the same value for the only free parameter. Its mean value is  $\gamma = 1.50 \pm 0.10$ . We note here that at an internuclear distance of  $a = 1.5a_0Z^{-1}$ the energy difference between the continuum and the molecular ground state is about  $E = 2.5Z^2$  Ry (1 Ry = 13.6 eV). In all cases the first and most pronounced anisotropy peak appears at about this energy. This suggests that, similar to the case of the atomic radiative-electron-capture effect,<sup>10</sup> transitions from the continuum into the split molecular ground state might be responsible for the observed effect. The energy of these transitions can exceed even the K ionization energy in the corresponding united atom, a fact which would explain why the periodic structure extends even beyond this limit.

Summarizing one can say that the suggested

model describes the kinematics of the observed anisotropy structure. But only a detailed investigation of the dynamics of radiative transitions in a rotating two-center Coulomb potential will show whether this model can explain the transition rates and the fact that this transition probability is enhanced exactly at the evaluated internuclear distance.

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