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Spectral Shape and Cross Section of Molecular-Orbital X-Ray Continua from Heavy-Ion Collisions

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We investigate molecular-orbital (MO) x-ray transitions resulting from collisions between 7–95-MeV sulfur ions and various thin, solid targets. MO radiation profiles beyond the united-atom limit are measured over several orders of magnitude in intensity, and absolute contributions of spontaneous and induced MO x rays are identified. We discuss formulations of MO theories which are in excellent agreement with our experimental spectral shapes and cross sections.

Observation of x rays from heavy-ion collisions has become an important tool to aid our still limited understanding of complex phenomena which occur in heavy-ion encounters. Careful measurements of collision-induced x rays have revealed that characteristic x-ray or satellite lines are always accompanied by relatively weak but distinct radiation continua. Almost at the same time, two mechanisms have been proposed to explain the production of such continua, molecular-orbital (MO)[1] and radiative-electron-capture (REC)[2,3] transitions. Since then, extensive further work has been reported especially on MO phenomena, but important features of this process such as precise spectral shapes and production cross sections remain poorly explained. It is the purpose of this Letter to clarify some experimental and theoretical questions on these radiation continua. In particular, we demonstrate that (i) MO and REC transitions can occur simultaneously in heavy-ion collisions and may give overlapping contributions to radiation continua, (ii) spectral shapes near and beyond the transition energy of the united (stationary) system, E_u, can be derived from a clarified formulation of the theory by Macek and Briggs[4] and are found to agree with experimental MO profiles over several orders of magnitude in intensity, and (iii) absolute MO production cross sections can be understood if one takes into account spontaneous and induced transitions.[5] We confine ourselves to the decay of K-shell vacancies and concentrate on MO tails beyond E_u which, in most cases, represent only a small though very distinct fraction of the total MO intensity. Our calculations for systems which need not be symmetric do not require detailed knowledge of the MO level diagram, and extension to initial vacancies in

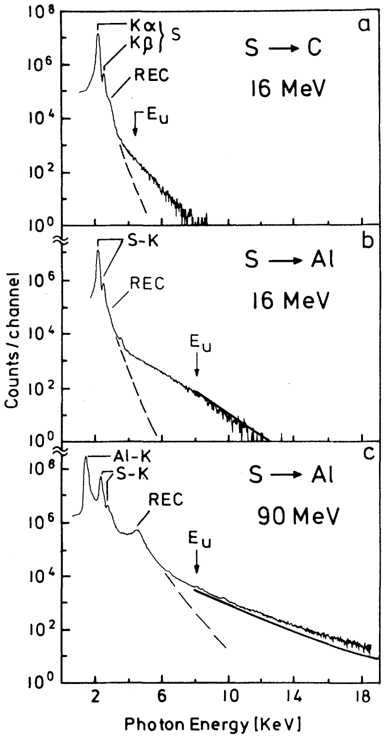

FIG. 1. X-ray spectra induced by sulfur ions impinging on 100-μg/cm^2 C and Al targets, corrected for absorber effects. The width of one channel is 18.2 eV. Dashed lines: calculated REC tail (Ref. 6); solid lines: MO tail calculated from Eqs. (1) and (2) for S→Al. Note virtual absence of background radiation.

L and higher shells seems possible.

Sulfur ions with energies between 7 and 95 MeV from the tandem Van de Graaff accelerator of the University of Munich were directed onto self-supporting foil targets (especially C and Al) with thicknesses between 5 and 200 μg/cm^2. X rays were observed with an 80-mm^2 Si(Li) detector with a resolution of 165-eV full width at half-maximum at 5.9 keV, placed at 90° to the beam direction. Use of various absorbers allowed us to obtain spectral intensities of both weak radiation tails and the relevant characteristic lines. Some results are shown in Fig. 1. The systems studied were chosen such that neither characteristic lines nor implantation and recoil effects influenced the line tails under investigation. Furthermore, our spectra are free of disturbing backgrounds. Calculation of absolute differential cross sections for electron and nuclear dipole and quadrupole bremsstrahlung showed that these effects are indeed orders of magnitude too small to have any measurable influence on our data.

Let us first elucidate the mechanisms which lead to MO and REC continua. There is a finite probability that a vacancy in a projectile ion decays by a radiative transition during an encounter with a target atom. On the one side, when the intrinsic velocity v_e of the jumping electron is large compared to the collision velocity v, molecular orbitals are formed and MO x rays result. On the other side, slow target electrons with $v_e < v$ can also undergo a dipole transition into the projectile vacancy and REC x rays are emitted. Thus, for beam velocities $v \lesssim v_e$ MO radiation dominates, but as soon as v exceeds v_e, REC processes involving outer target electrons become important and will generally dominate the radiation continuum near the characteristic projectile x-ray line. A theoretical description of REC phenomena has been published recently[7]; we have significantly refined this model in order to separate proper MO contributions in the present experimental x-ray spectra. A detailed and extensive description of experimental and theoretical REC results is in preparation.[6,8]

There has been some confusion with respect to the true spectral shape of MO x rays. Many authors reported pronounced peaks of MO radiation[1] and associated certain half-widths to these peaks though it appears now that these effects, at least for K- and L-shell transitions, are simply caused by absorbers. Identification of MO tails often rested on the alleged observation of a cutoff[9] near E_u; however, unambiguously clean cutoffs have never been presented and theoretical considerations have revealed that sharp cutoffs cannot be expected. Below, we investigate the MO x-ray profile in that critical energy range near and beyond E_u.

The most advanced treatment of MO tails was given by Macek and Briggs.[4] In their view, the observed x-ray intensity is composed of two parts, (i) the actual MO contribution and (ii) a Lorentzian tail of the undistorted characteristic x-ray line, whereby (ii) dominates (i) for x-ray energies E_x which are slightly above E_u. For two reasons, we argue that contribution (ii) must be dropped: Firstly, one cannot assume that a natural line of width Γ, centered at E_0, can be described by a Lorentzian distribution for $E_x - E_0 \gg \Gamma$ [note $(E_x - E_0)/\Gamma > 10^3$]. Secondly, there must be a sharp cutoff for transition energies which exceed the binding energy of the final projectile state in question. Still larger excitation energies would involve initial states which are strongly coupled to the continuum; we do not

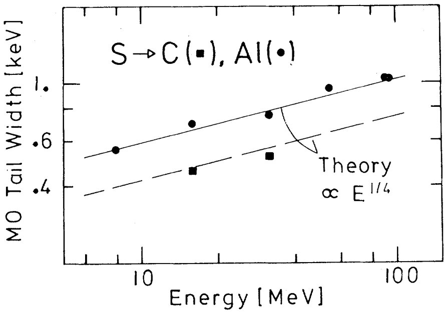

FIG. 2. Half-width of MO tails near the united-atom energy, as a function of sulfur projectile energy. Theoretical curves are calculated from Eq. (3). Experimental errors are of the order of the size of the data points.

question that such states may be produced, but if they are the result of a heavy-ion collision it is likely that the dominant part of electrons excited into these states of a then isolated ion will simply escape instead of giving rise to a dipole transition with release of extraneous energy. For impact energies which are high enough to allow sufficient shell interpenetration, we estimate the proper two-collision MO cross section for spontaneous transitions with energies beyond the united-atom limit, $E_x \gtrsim E_u$, by means of the saddle-point method and obtain, in extended analogy with Ref. 4,

$$\frac{d\sigma^{\mathrm{sp}}}{dE_x} \simeq \frac{2\pi R_u{}^2 g^2(y) E_x}{\hbar \tau_u \dot{\omega} E_u}; \quad y = \frac{E_x - E_u}{\hbar(\pi\dot{\omega})^{1/2}}, \tag{1}$$

where R_u is the K-shell radius of the united atom, $\tau_u{}^{-1}$ is the radiative transition rate for the united atom, g is a dimensionless standard function involving Fresnel integrals,[10] and $\hbar\dot{\omega} = v\Delta E_x/\Delta R$ approximates the average change of transition energy with varying internuclear separation R of the collision partners.

Radiative transitions can also be induced as a result of the rotation of internuclear axis in close collisions.[5] The corresponding MO cross section can be evaluated for $E_x \gtrsim E_u$ and yields, when x rays are observed at 90° to the beam direction,

$$\frac{d\sigma^{\mathrm{ind}}}{dE_x} \simeq \frac{d\sigma^{\mathrm{sp}}}{dE_x}\,\frac{9}{8}\left(\frac{\hbar v}{R_u E_u}\right)^2 \ln\left(\frac{R_u}{R_m}\right), \quad E_x \gtrsim E_u, \tag{2}$$

where R_m denotes the minimum distance of closest approach in a head-on collision.

According to Eq. (1), the intensity near and beyond E_u decreases nearly exponentially with a half-width given by

$$H \simeq 0.3\hbar\dot{\omega}^{1/2} = 0.3(\hbar v\,\Delta E_x/\Delta R)^{1/2}, \tag{3}$$

where we approximate ΔE_x by the difference of transition energies for separated and united ion and ΔR by the relevant sum of separated shell radii, $R \simeq R_1(1 + Z_2/Z_1)$ with $Z_1 \geq Z_2$. The resulting widths closely reproduce the experimental ones (Fig. 2). Equation (3) can also be used to describe tails recently communicated for Ni → Ni collisions.[11] The result $H \propto v^{1/2}$ should hold for both spontaneous and induced transitions; the assumption of a Heisenberg-type broadening alone,[5] $H \propto v$, is clearly at variance with both Eq. (3) and experimental data.

Convenient comparison between experimental and theoretical absolute production of MO x rays with energies $E_x \geq E_u$ becomes possible when one integrates Eqs. (1) and (2) over dE_x for $E_x \geq E_u$ and normalizes to the associated characteristic projectile x rays; this gives the fractions

$$f^{\mathrm{sp}} \simeq 0.22\pi N R_u{}^2(\hbar v\,\Delta R/\Delta E_x)^{1/2}\tau_0/\tau_u, \tag{4}$$

$$f^{\mathrm{ind}} \simeq 0.76 N\tau_0\tau_u{}^{-1}E_u{}^{-2}(\hbar v)^{5/2} \times (\Delta R/\Delta E_x)^{1/2}\ln(R_u/R_m), \tag{5}$$

where N is the density of target atoms and τ_0 represents the radiative lifetime of the vacancy in the isolated projectile ion. We evaluated f^{sp} and f^{ind} for our system S + Al and obtain ratios which are shown in Fig. 3 along with the experimental data. To some extent, the resulting close agreement between theoretical estimates (with no freely adjustable parameter) and measured fractions may be fortuitous. Still, it appears that our results prove the importance of induced emission. Previous attempts along these lines[11, 12] are not supported since for the cases reported there Eq. (2) predicts no appreciable influence of induced transitions.

Finally, we give some approximate scaling laws for symmetric collisions ($Z_1 = Z_2 = Z$). Absolute MO-tail cross sections from Eq. (2) become $\sigma^{\mathrm{sp}} \simeq 0.47 \times 10^{-6}\pi a_0{}^2(Zv_0/v)^{1/2}$, where $v_0 = e^2/\hbar$. Equations (4) and (5) yield $f^{\mathrm{sp}} \simeq 0.15 Z^{-7/2} \times (E/A)^{1/4}$, where E and A denote projectile energy in MeV and mass number, respectively, and $f^{\mathrm{ind}} \simeq 8Z^{-11/2}(E/A)^{5/4}\ln(R_u/R_m)$. Equality of spontaneous and induced emission occurs for impact energy $E' \simeq (Z^3/121)$ MeV. We note, however, that our derivations are based on a two-collision MO production mechanism. Especially for higher Z, one-collision processes, production and decay of a vacancy during a single collision, become im-

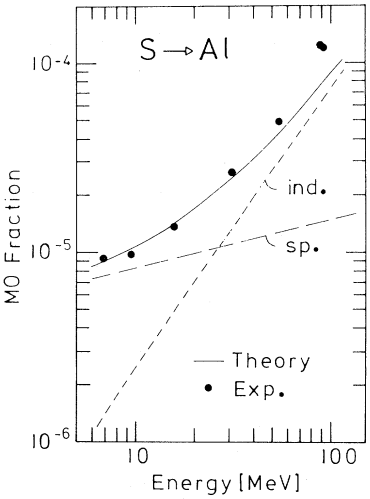

FIG. 3. X-ray intensity ratio between MO tail beyond the united-atom energy and S K line, as a function of sulfur projectile energy. Contributions from spontaneous (sp) and induced (ind) transitions and the sum of both (solid line) are indicated as calculated from Eqs. (4) and (5).

portant. From experiments with gas targets we have attained enlightening results on this very important question; these will be communicated elsewhere.

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Light Scattering through the Isotropic-Cholesteric Phase Transition of a Cholesteric Liquid Crystal

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We have measured the Rayleigh intensity and linewidth in cholesteryl oleyl carbonate continuously through the cholesteric-isotropic phase transition. Above the phase transition the intensity data do not agree with the theory of de Gennes; we also do not observe the instability of the isotropic phase predicted by de Gennes. At the phase transition we observe *two* exponentials simultaneously which we associate with order-parameter and director fluctuations.

Despite extensive measurements on director fluctuations in nematic phases[1,2] and order-parameter fluctuations in isotropic phases[3-7] of liquid crystals, it has so far been impossible to observe the passage from one type of fluctuation to the other because of the presence of a weak first-order transition separating the two phases. Indeed, de Gennes[8] has recommended looking for liquid crystals with "metastable phases" for just this purpose.

In this Letter we report measurements of the Rayleigh intensity and linewidth of cholesteryl oleyl carbonate (COC), a cholesteric liquid crystal which appears to have the metastable state mentioned by de Gennes. We summarize our observations as follows:

(1) The optical properties of COC change *continuously* upon cooling from the isotropic phase to the cholesteric phase without a detectable first-order phase transition.