Infrared Divergence of the Resonant Raman-Compton Scattering

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The infrared divergence of the resonant Raman-Compton scattering has been studied in collisions of photons on atomic L electrons in the intermediate-momentum-transfer regime. Low-energy continua emitted by Zr atoms, excited, in the vicinity of the K edge, by the monochromatized x rays delivered by the LURE Synchrotron Radiation Facility, have been observed on very thin targets and compared with the theoretically predicted infrared divergence of the Raman scattering. The characteristic change in shape of these continua has been studied over a wide energy range below the Zr K edge.

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While the Raman scattering in molecules is a well-known effect and a well-established technique, this process has only very recently be considered in the x-ray regime where inner-core excitations are involved. In such a case, where autoionizing levels are considered, this process exhibits a very different behavior, allowing the study of new effects through some yet unconsidered terms of the theory. In this process, schematically described in Fig. 1, an incoming photon $h\nu_i$ is inelastically scattered by an atom, which is excited in the final state of the collision. When the intermediate (virtual) state V is close to a discrete (real) level, this process is resonant. When considering inner-core excitation, two decay processes of the virtual level may be taken into account: (a) The first is the well-known Raman scattering [Fig. 1(a)] where the target atom is excited, and the incoming photon scattered at a lower energy. This process, in which a monoenergetic scattered line is emitted, has only quite recently been observed on an atomic discrete level.[1] This process is resonant on both sides of the discrete level; in the final state the atom is excited, with an L electron being promoted to an empty outermost level. (b) The second is the Raman-Compton scattering [Fig. 1(b)] where, in the decay channel, the available energy is shared between a photon and an electron (autoionizing level); continuum photon (and electron) energy spectra are then observed, whose maximum energy edges are equal to the total available energy ($\approx h\nu_i - B_L$). This process is then asymmetrically resonant near the ionization energy level (the energy difference between the virtual level and the $1s$ ionization limit will be denoted by ϵ), and in the final state the atom is ionized in the L shell.

The virtual level decays by either (a) or (b). In the case of solid targets where the outermost excited electrons are not located in discrete energy states but in broad bands, these two processes cannot be experimentally separated and only complex continuum spectra are observed.

In the example considered in Fig. 1, which is among the most typical cases, the real resonant state corresponds to the excitation of a $1s$ electron to an np unoccupied electronic state of the atom $(1s)^{-1}(np)^{+1}$ while the final state is made of a $2p$ electron excited in this np state: $(2p)^{-1}(np)^{+1}$. The maximum energy of the Raman-Compton continuum is then equal to $h\nu_i - B_L = K\alpha - \epsilon$. The resonant character of this process has been first demonstrated by Sparks[2] on solid targets where processes (a) and (b) cannot be separated. The two processes have only recently been separately ob-

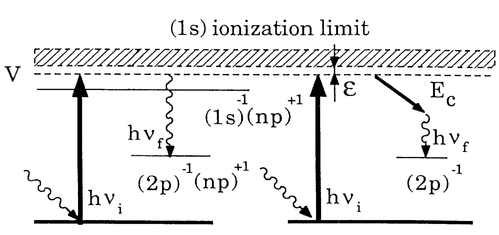

FIG. 1. Resonant Raman-Compton effect: (a) Raman scattering (inelastic scattering of a photon) and (b) Raman-Compton scattering (the energy given to the atom is, in the final state, shared between an electron and a photon).

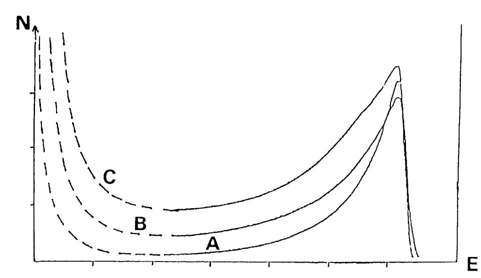

FIG. 2. Theoretical (energy normalized) photon spectra of Raman-Compton scattering plus infrared divergence (dashed line) from Ref. 3 showing the dramatic change in shape for various relative detuning energies $\epsilon/h\nu_i = 0.12$ (curve A), 0.20 (curve B), and 0.38 (curve C). (The relative intensities of these spectra are arbitrary.) The dashed lines are the theory of Gavrila and Tugulea (Ref. 4).

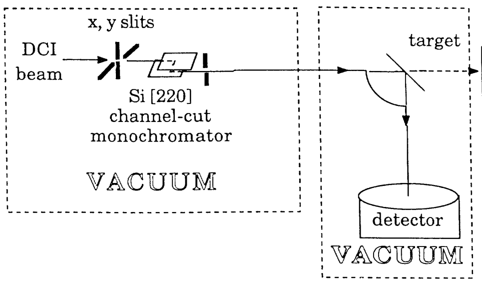

FIG. 3. Schematic of the design of the experiment (on the high-energy storage ring of Orsay DCI dedicated to synchrotron radiation).

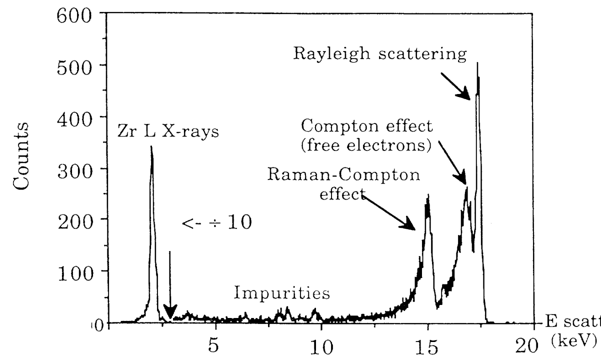

FIG. 4. Typical scattered spectrum at $h\nu_i = 17.4$ keV.

served by Braind *et al.*,[1] as well as their two different resonant behaviors (on the discrete and on the continuum spectra). The two ejected particles (scattered photon and extracted electron) share in a continuous way the available energy. The high-energy part of the continuous photon spectrum appears as an edge followed at lower energy by a plateau, observed by Bennett, Rapoport, and Freund[3] (Fig. 2). In 1972, Gavrila[4,5] predicted the existence of a rise of this plateau in the lower-energy side of the spectrum (infrared divergence of the Raman-Compton scattering) (Fig. 2). The infrared divergence is a very general effect in many problems in physics when scattered particles have energies going to zero. Interesting physical insight on infrared divergences will be found in some review papers in Refs. 6 and 7, e.g., the correspondence between infrared divergence in Raman-Compton scattering and fluorescence at energies close to zero. As predicted by Gavrila and Tugulea,[4,5] the intensity of this infrared divergence as well as its "width" must theoretically evolve in a similar way to the Raman-Compton edge as a function of the incoming photon energy. A typical evolution of the whole spectrum is presented in Fig. 2 which shows how dramatic is the change in shape of these continua as a function of the energy of the incoming photon. The purpose of this Letter is to study this infrared divergence.

The main problem to solve when observing such a low-energy continuum is to get rid of all the processes which usually provide low-energy background in the target as well as in the detector. Because of the small value of the Raman-Compton scattering cross section compared to other interaction processes of photons on atoms (namely the dominant one, L photoionization), all the experiments carried out so far used very thick targets as in Refs. 3, 8, and 9. We have then chosen to study this process by using very thin (few tenths of μm) targets irradiated by an intense source of monochromatized x rays. The design of the experiment is presented in Fig. 3.

The x rays were provided by the synchrotron light delivered by the Laboratoire pour l'Utilisation du Rayonnement Electromagnétique (LURE) Synchrotron Radiation Facility in Orsay. The continuous x-ray spectrum was monochromatized with a Si[220] channel-cut crystal which allowed the production of a continuously tunable x-ray beam of energy between a few keV up to 19 keV (maximum available energy at LURE with a reasonable intensity) within a few eV bandpass. This beam was incident on very thin samples of zirconium (the heaviest available element, whose K binding energy is close to 18 keV). The scattered photons were detected at 90° with a SiLi detector of 155-eV resolution at 6 keV, and whose beryllium window entrance was thin enough (25 μm) to allow the detection of photons down to 1.5 keV and whose energy transmission and efficiency were carefully measured.[10]

We present in Fig. 4 a typical energy spectrum observed close to the resonance at 17.4 keV (0.6 keV below the K ionization energy), where most of the main interaction processes are seen: Rayleigh scattering (at the incoming energy), Compton scattering characteristic of the lightly bound electrons, some fluorescence lines corresponding to the unremovable impurities in zirconium (< 100 ppm), and the characteristic L lines of zirconium corresponding to the main interaction processes (by orders of magnitude) for photons of energy lower than the K binding energy, i.e., the L ionization. The high-energy edge of the Raman-Compton scattering can also be seen

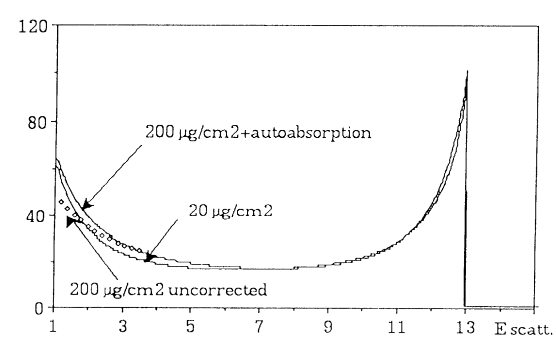

FIG. 5. Comparison of low-energy continua observed with thick and thin targets.

at an energy equal to $h\nu_i - B_L$. The L x rays have been subtracted first from the experimental spectra in the following way. The L characteristic lines of Zr following L photoionization have exactly the same shape at all impinging energies but different intensities (power of -3 scaling law). These lines have been smoothed and subtracted from the experimental spectra by using exactly the same procedure at every energy by conventional fitting procedures. As shown in Fig. 6 this procedure leads to the appearance of very different shapes for these observed continua from one excitation energy to another one, a fact which cannot be explained by any of the processes described below, when the photon energies are tuned in such a narrow energy range (14–17 keV).

As previously quoted, the low-energy continuous spectra can be interpreted as background coming from the detector and/or the target. The continuous tails usually observed in SiLi detectors irradiated by monochromatic

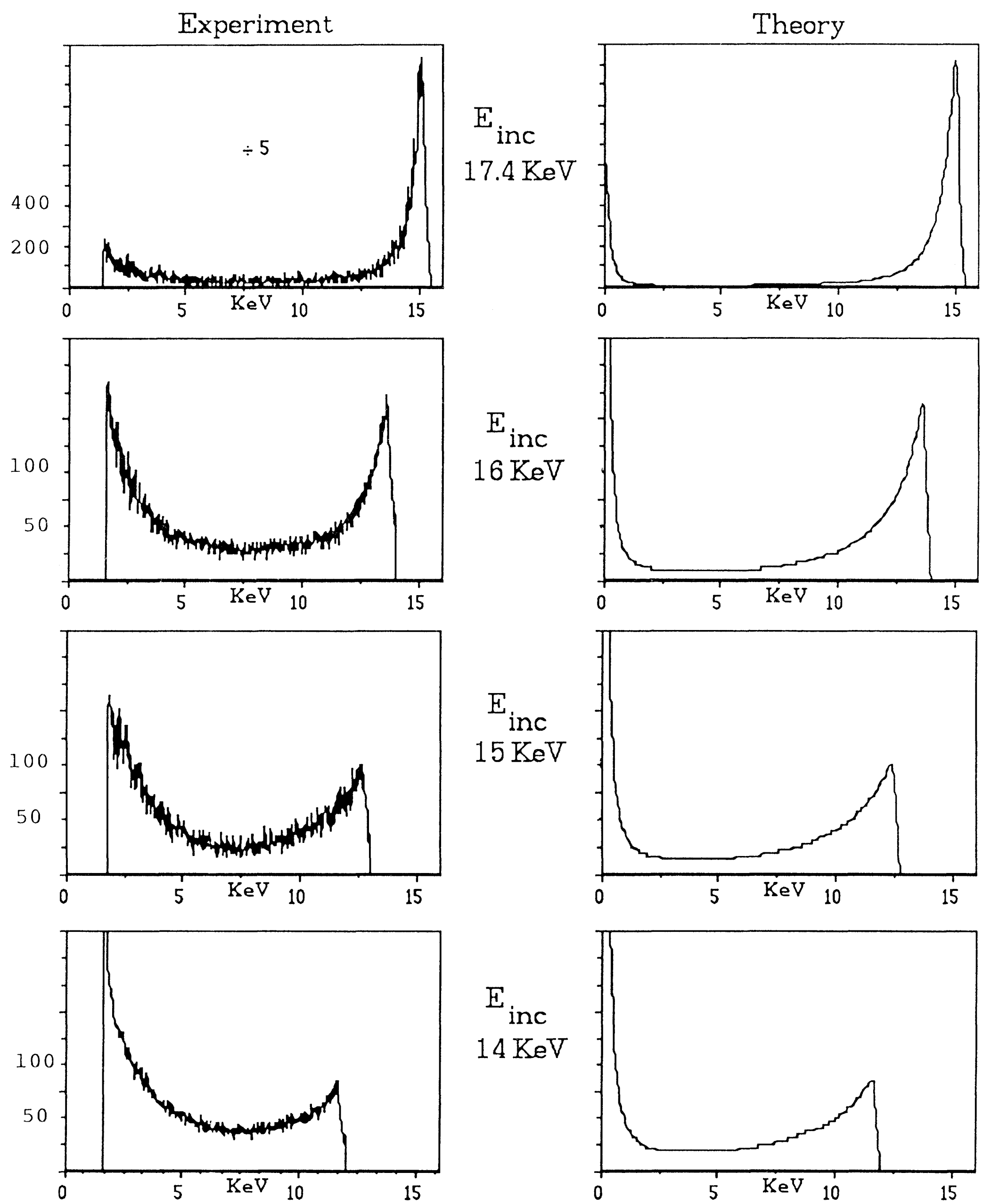

FIG. 6. Raman-Compton spectra at various energies: left-hand panels, experimental data after removing L x-ray fluorescence; right-hand panels, theoretical predictions normalized on the high-energy profile.

radiation, which mainly correspond to incomplete charge collection (and which are usually flat), have been studied at seven energies ranging from 3 to 13 keV. These experiments, carried out with fluorescence techniques induced by synchrotron radiation, allowed us to subtract from the experimental spectra all the lines or continua, with the exception of the Raman-Compton profile under study, with their own (detector included) low-energy backgrounds. The residual spectrum which then looks like a bowl-shaped spectrum, similar to that predicted by the theory for the Raman-Compton spectrum (high-energy profile plus infrared divergence), has then only to be studied from the viewpoint of backgrounds induced in the target.

The major cause of background to be considered in the foil comes from the main interaction of the photons with the atomic target, i.e., L photoionization. This process, whose cross section is 2 or 3 orders of magnitude larger than the Raman scattering, provides a large number of photoelectrons ($E_e = h\nu_i - B_L \sim 10$ to 16 keV in the considered experiment) that may slow down in the foil giving rise to bremsstrahlung photons with a characteristic spectrum very similar to that of the infrared divergence. In order to estimate the contribution of this process we have studied the characteristic low-energy x-ray spectra observed with various targets (and target backings) of several thicknesses. We used very thin Zr targets such that the range of 12-keV photoelectrons (380 $\mu g\,cm^{-2}$) is much larger than the thickness of the target (20 $\mu g\,cm^{-2}$). We can expect in these targets a very small or negligible bremsstrahlung as compared to the infrared divergence. We then compared the spectra to those obtained with thicker targets ranging up to 200 $\mu g\,cm^{-2}$ and only found a linear dependence between the spectrum intensity and the target thickness instead of the quadratic one that would originate from some bremsstrahlung processes. We present in Fig. 5 a comparison of two extreme spectra showing that, even for thick targets, the contribution of bremsstrahlung from L photoelectrons is negligible (a fact in agreement with calculations[3] but which needed to be experimentally demonstrated). To record a spectrum with very thin targets (for which autoabsorption corrections are not necessary) the exposure is of about 10 to 15 h (same order of magnitude as the lifetime of the positron beam in the synchrotron storage ring). We then studied the Raman scattering on very thin targets (20 $\mu g/cm^2$) only for two different energies, 15 and 16 keV, and on thicker ones for the others.

The corrected spectra observed at different energies, ranging from 14 to 17.4 keV ($B_K = 18$ keV) are presented in Fig. 6 and compared to the theoretical predictions of the infrared divergence. Low-energy continua have been unambiguously observed, whose dramatic changes in shape and intensity follow qualitatively those of Raman-Compton profiles, a fact which cannot be attributed, as discussed above, to any bremsstrahlung or experimental effects. These continua, whose absolute intensities are in rough agreement with theory, are, however, much broader than theoretically predicted, as in the case of K scattering[8,9] for thick targets, a fact that cannot be explained for the moment, and that cannot be attributed to any outermost-shell effect (M) whose intensity has been previously studied by Kodre and Shafroth.[10] Details on theoretical calculations, polarization effects, absolute cross-section measurements, and the effect of outermost electrons will be soon presented in a more exhaustive paper on Raman-Compton scattering.[11]

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[1]J. P. Briand, D. Girard, V. Kostroun, P. Chevallier, K. Wohrer, and J. P. Mossé, Phys. Rev. Lett. **46**, 1625 (1981).

[2]C. J. Sparks, Jr., Phys. Rev. Lett. **33**, 262 (1974).

[3]Y. B. Bannett, C. Rapoport, and I. Freund, Phys. Rev. A **16**, 2011 (1977).

[4]M. Gavrila and M. N. Tugulea, Rev. Roum. Phys. **20**, 209 (1975).

[5]M. Gavrila, Phys. Rev. A **6**, 1348 (1972).

[6]T. Åberg and J. Tulkki, in *Atomic Inner-Shell Physics*, edited by B. Crasemann (Plenum, New York, 1985).

[7]L. Rosenberg, Adv. At. Mol. Phys. **18**, 1 (1982).

[8]G. Basavaraju, P. P. Kane, and Suju M. George, Phys. Rev. A **36**, 655 (1987).

[9]V. Marchetti and C. Franck, Phys. Rev. A **39**, 647 (1989).

[10]A. F. Kodre and S. M. Shafroth, Phys. Rev. A **19**, 675 (1979); see also A. F. Kodre *et al.*, Z. Phys. A **297**, 25 (1980).

[11]A. Simionovici, thesis, University of Paris, 1988 (unpublished); A. Simionovici *et al.* (to be published).