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Geant4 implementation of inter-atomic interference effect in Small-Angle Coherent X-ray Scattering for materials of medical interest

G. Paternò, P. Cardarelli, A. Contillo, M. Gambaccini and A. Taibi

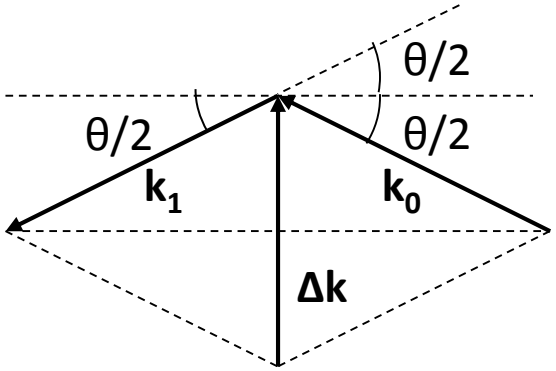
Dipartimento di Fisica e Scienze della terra e INFN - Ferrara

Outline

- Theoretical background
- Implementation in Geant4
- Case studies

Theoretical background: Coherent Scattering

In **Rayleigh (Coherent) Scattering**, photons are scattered by **bound atomic electrons** without excitation of the target atom, i. e., the energy of incident and scattered photons is the same.



$$\frac{d\sigma_{Ra}}{d\Omega} = \frac{d\sigma_{Th}}{d\Omega}\left|F(q,Z) + f' + if''\right|^2 \approx r_e^2 \frac{1+\cos^2\theta}{2}\left|F(q,Z)\right|^2$$

Dispersion correction, negligible for materials and energies of medical interest (we are above K absorption edges)

momentum transfer

$$q = \eta\,|\,k_1 - k_0\,| = 2k\sin(\theta/2) = \frac{4\pi}{\lambda}\sin(\theta/2) = 2\frac{E}{c}\sin(\theta/2)$$

Parameters used in the literature and MC codes

$$x = \frac{q}{2h} = \frac{1}{\lambda}\sin(\theta/2) \quad [\text{nm}^{-1}] \qquad \tilde{q} = \frac{q}{m_e c} \quad [\text{adimensional}]$$

$$\sigma_{Ra} = \int \frac{d\sigma_{Ra}}{d\Omega}\,d\Omega = \pi r_e^2 \int_0^{\pi} (1+\cos^2\theta)\left|F(q,Z)\right|^2 \sin\theta\, d\theta$$

For low photon energies: σ_{Ra} 🕓 $\sigma_{Th} = 8/3\pi r_e^2 Z^2$

For high photon energies (E>Z/2 MeV): $\sigma_{Ra} \sim E^{-2}$

Theoretical background: Atomic Form Factor

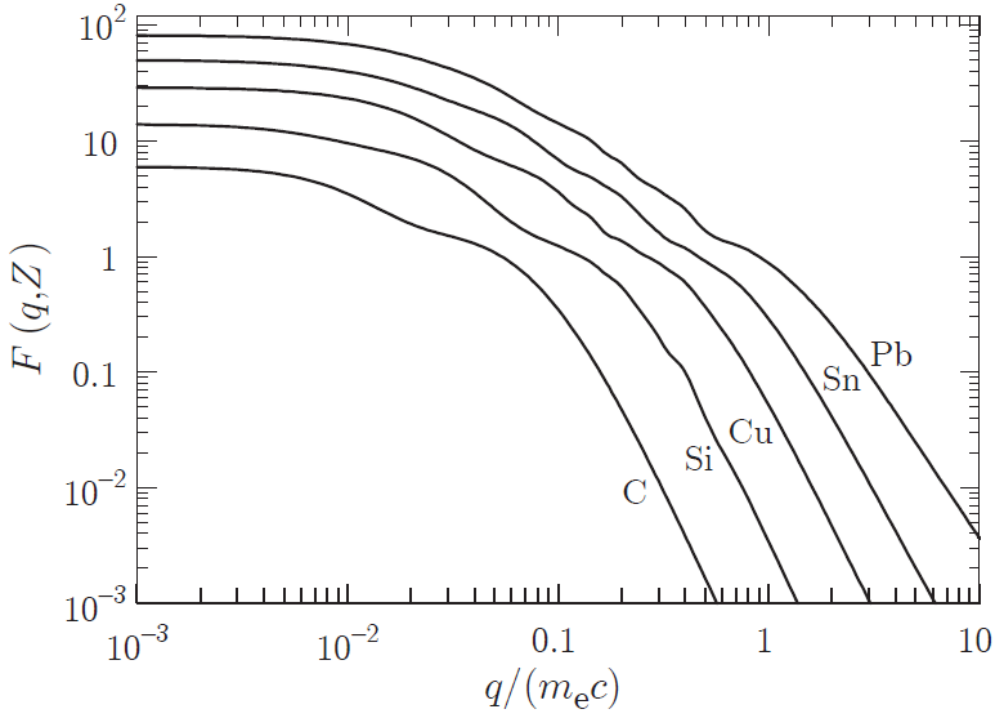
The atomic form factor, F(q,Z) is the Fourier transform of the atomic electron density ρ(r).

For spherically symmetric atoms:

$$F(q,Z) = 4\pi \int_0^\infty \rho(r) \frac{\sin(qr/\eta)}{qr/\eta} r^2 dr$$

F(q,Z) is a **monotonically decreasing** function of q that varies from **F(0,Z) = Z to F(∞,Z) = 0,** thus resulting **in a forward peaked scatter distribution**.

The most accurate form factors are those obtained from **non-relativistic Hartree-Fock calculations** (see, **Hubbell et al., 1975)** on which is based **EPDL97** of **LLNL**).



Atomic form factors of neutral atoms of the indicated elements, taken from the EPDL (Cullen et al., 1997).

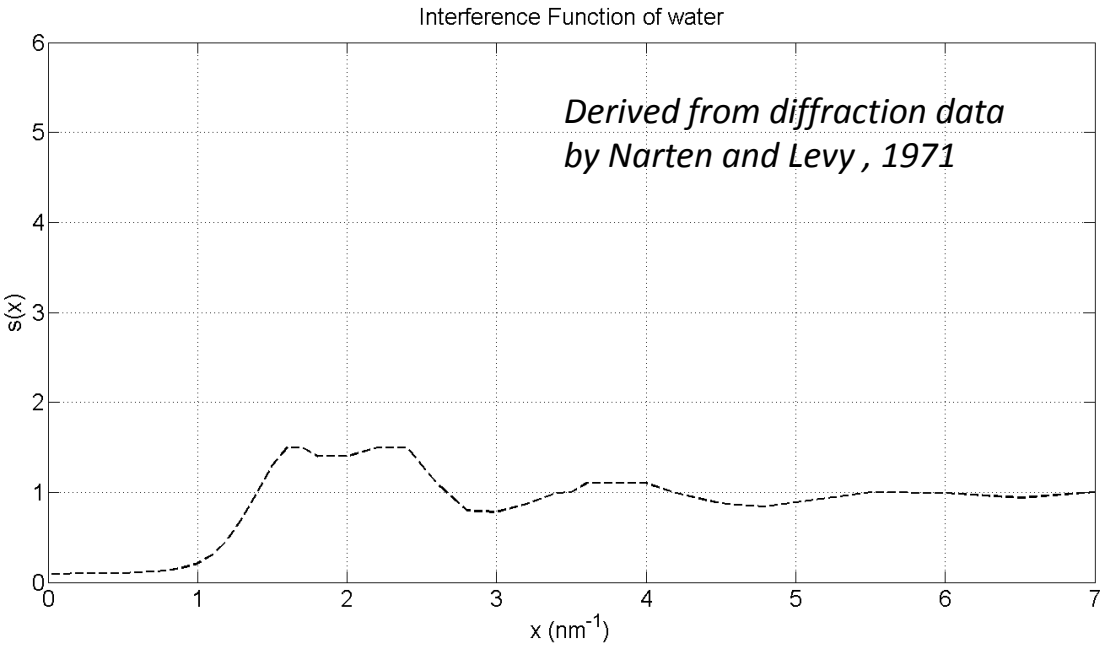
Theoretical background: Molecular Form Factor

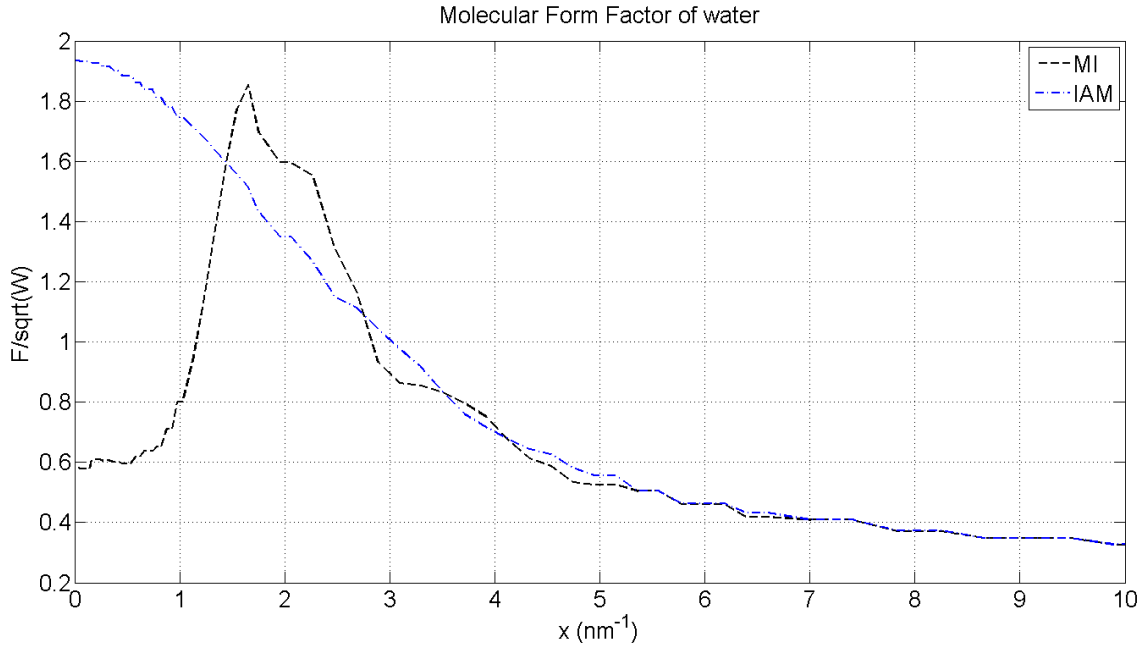
$$F^2_{mol,IAM}(q) = W\sum_i \frac{w_i}{A_i} F^2(q, Z_i)$$

$$F^2_{mol}(q) = F^2_{mol,IAM}(q)\cdot s(q)$$

Independent-Atoms Model (IAM)

Molecular Interference (MI) effects appear in **liquid** and **amorphous solids** (not only in crystals). **The Interference function s(q)** depends on the statistical arrangement of the molecules (**short-range order**). It shows an **oscillatory behavior** and **tends to unity at high q values**. **It depends on the material** and is derived from **XRD experiments**.





The fraction of coherent scattering interactions is about 10% for materials and energies of medical interest but, due to MI, coherent radiation is not forward peaked and is distinguishable from primary radiation.

Geant4 simulation toolkit

Geant4 is a **open-source C++ based object-oriented** Monte Carlo toolkit for particle transport in matter. It is routinely used in **many scientific disciplines** included medical science. It provides:

- advanced geometry modeling,
- high quality physics models,
- advanced tracking algorithms,
- interactive facilities for visualization and execution.

For each physical process various models are available (specialized for particle type and energy scope). **Electromagnetic** physics foresees **Standard** and **Low Energy** packages.

In **Standard models**, the energy of the particles > 1 keV, the atom nucleus is free, the atomic electrons are quasi-free, and matter is described as homogeneous, isotropic, amorphous.

The **Low Energy package** extends the coverage of electromagnetic interactions down to 250/100 eV, it includes process based on **detailed models** (atom shell structure, precise angular distributions, polarization, etc).

The **coherent scattering models** current implemented in the official release **do not take into account the influence of molecular interference** -> **scatter figure can not be rigorously evaluated**.

Electromagnetic physics in Geant4

EM processes → **Low Energy** →

in **two "flavors" of models**:
- based on the Livermore Library
- à la Penelope

- Multiple scattering
- Bremsstrahlung
- Ionization
- Annihilation
- Photoelectric effect
- Compton scattering
- Rayleigh scattering
- e+e- pair production
- Synchrotron radiation
- Transition radiation
- Cherenkov
- Scintillation
- Refraction
- Reflection
- Absorption
- Fluorescence
- Auger

Livermore

- Based on evaluated data libraries from LLNL (**mixture of experiments and theories for electrons and photons**):
  - EADL *(Evaluated Atomic Data Library)*
  - EEDL *(Evaluated Electrons Data Library)*
  - EPDL97 *(Evaluated Photons Data Library)*

  especially formatted for Geant4 distribution *(courtesy of D. Cullen, LLNL)*
- Validity range: 250 eV - 100 GeV
- Elements Z=1 to Z=100

Penelope

- The whole physics of Penelope code has been re-engineered into Geant4 (it benefit from OO power)
- Physics models by F. Salvat et al. (version 2008)
- **Mixed approach**: analytical, parameterized & data-driven (down to 100 eV)
- **Great care of atomic effects, fluorescence, Doppler broadening, etc**
- **Manages positrons**

MI effect implementation in Geant4

- **We modified Penelope model of Rayleigh scattering** (*G4PenelopeRayleighModel* class) in order to take into account MI effect by **reading custom molecular form factors** (through the new method: *ReadMolInterferenceData()*).

- **We prepared a database of modified molecular form factors for a set of material of medical interest** (various tissues and plastics). The files were positioned inside the directory "MIFF" located at the low energy data path:

  Geant4_installation_path/share/Geant4-10.3.1/data/G4EMLOW6.50/penelope/rayleigh/

- Modified molecular form factors can be accessed by **assigning proper names to the materials** used in the simulation.

List of implemented Molecular Form Factors

A total of 24 Molecular Form Factors have been included

Tartari et al., Phys. Med. Biol. 47 (2002), 163-175

- fat
- water
- collagen (bone matrix)
- hydroxyapatite (mineral)
- PMMA

Peplow and Verghese, Phys. Med. Biol. 43, No. 9 (1998), 2431-2452

- lucite, lexan, kapton, water
- pork heart, kidney, liver, muscle
- beef blood
- human breast

Kidane et al., Phys. Med. Biol. 44 (1999), 1791-1802

- carcinoma tissue

Chaparian et al., Iran. J. Radiat. Res., 2009; 7 (2): 113-117

- adipose
- glandular
- breast tissue (50% water - 50% lipid)
- water

Kosanevsky et al., Med. Phys. 14 (4) 1987, 527-532

- nylon
- polyethylene
- polystyrene
- gray matter

List of implemented Molecular Form Factors

tissue → { soft tissue (fat, water); bone tissue → { dry bone (mineral, non-mineral); red and yellow marrow (soft issue) }

A set of four components, namely **fat**, **water**, **bone matrix (BM)** and **hydroxyapatite (HA)**, **can represent a basis for the composition of the human tissues**. Once the basis is defined, **one can simulate any tissue by linear combination**.

$$F^2(q) = a_1 F^2_{fat}(q) + a_2 F^2_{water}(q) + a_3 F^2_{BM}(q) + a_4 F^2_{HA}(q)$$

Elemental composition by mass of the four basis materials

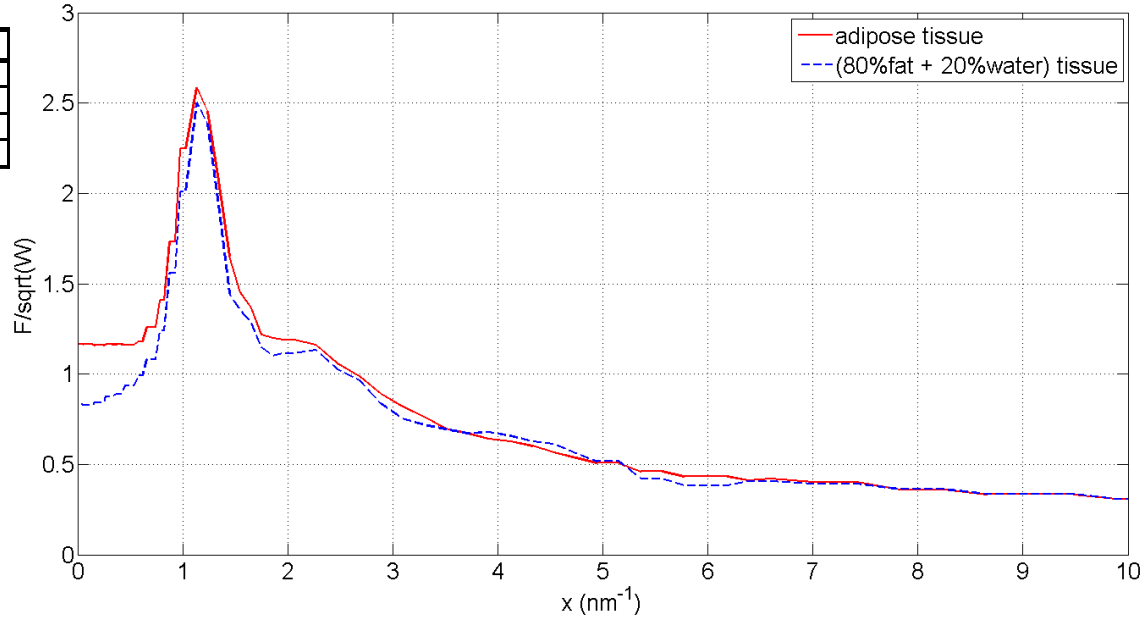
Substance	H	C	N	O	p	Ca	Density (g/cm^3)
water	0.1119			0.8881			1.00
fat	0.1190	0.7720		0.1090			0.923
bone matrix (collagen)	0.0344	0.7140	0.1827	0.0689			-
hydroxyapatite (mineral)	0.0020			0.4140	0.1850	0.3990	2.74

This approach was proposed in:

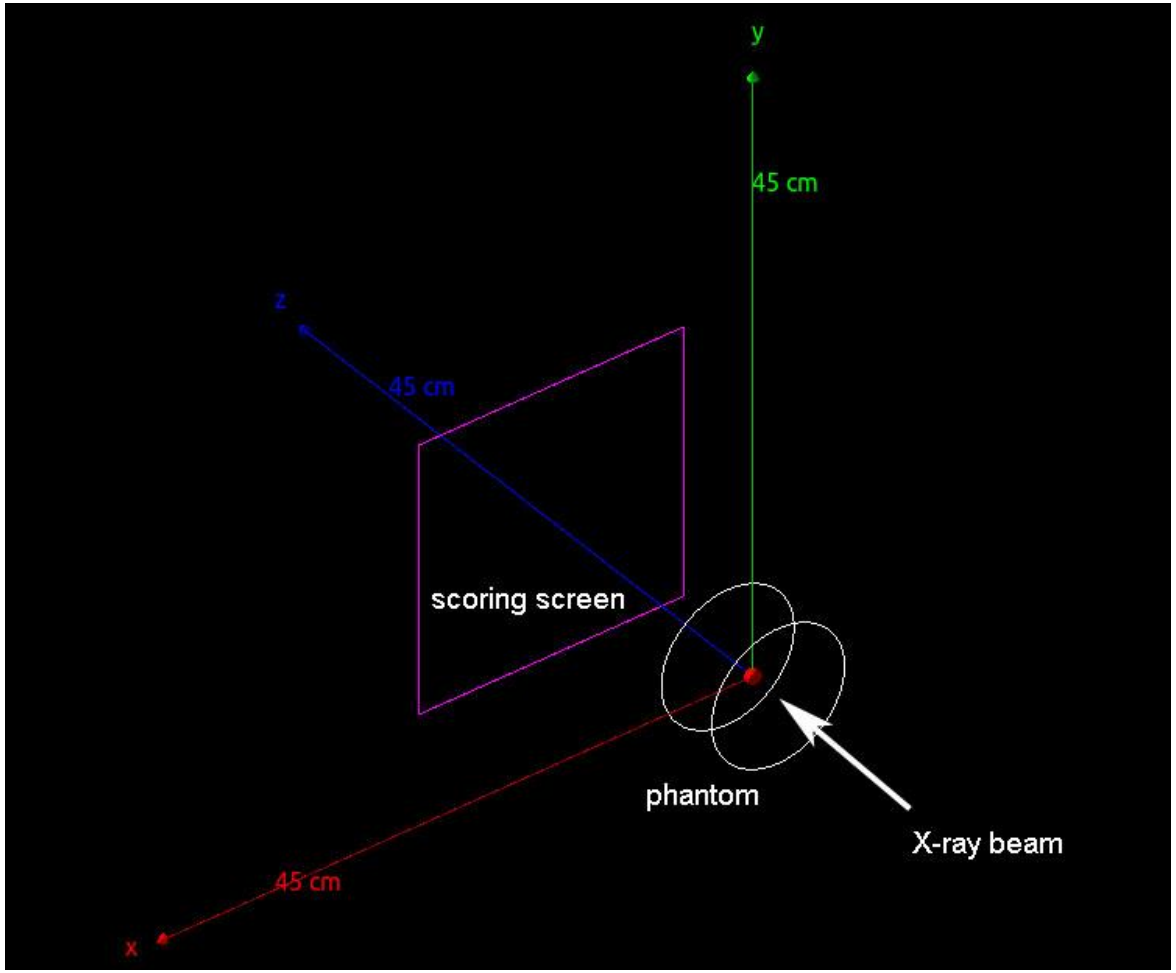
Taibi et al., Proceedings of the Monte Carlo 2000 Conference , Lisbon, 23–26 October 2000.

Tartari et al., Radiation Physics and Chemistry 61 (2001) 631–632.

Tartari et al., Phys. Med. Biol. 47 (2002), 163-175.



Validation

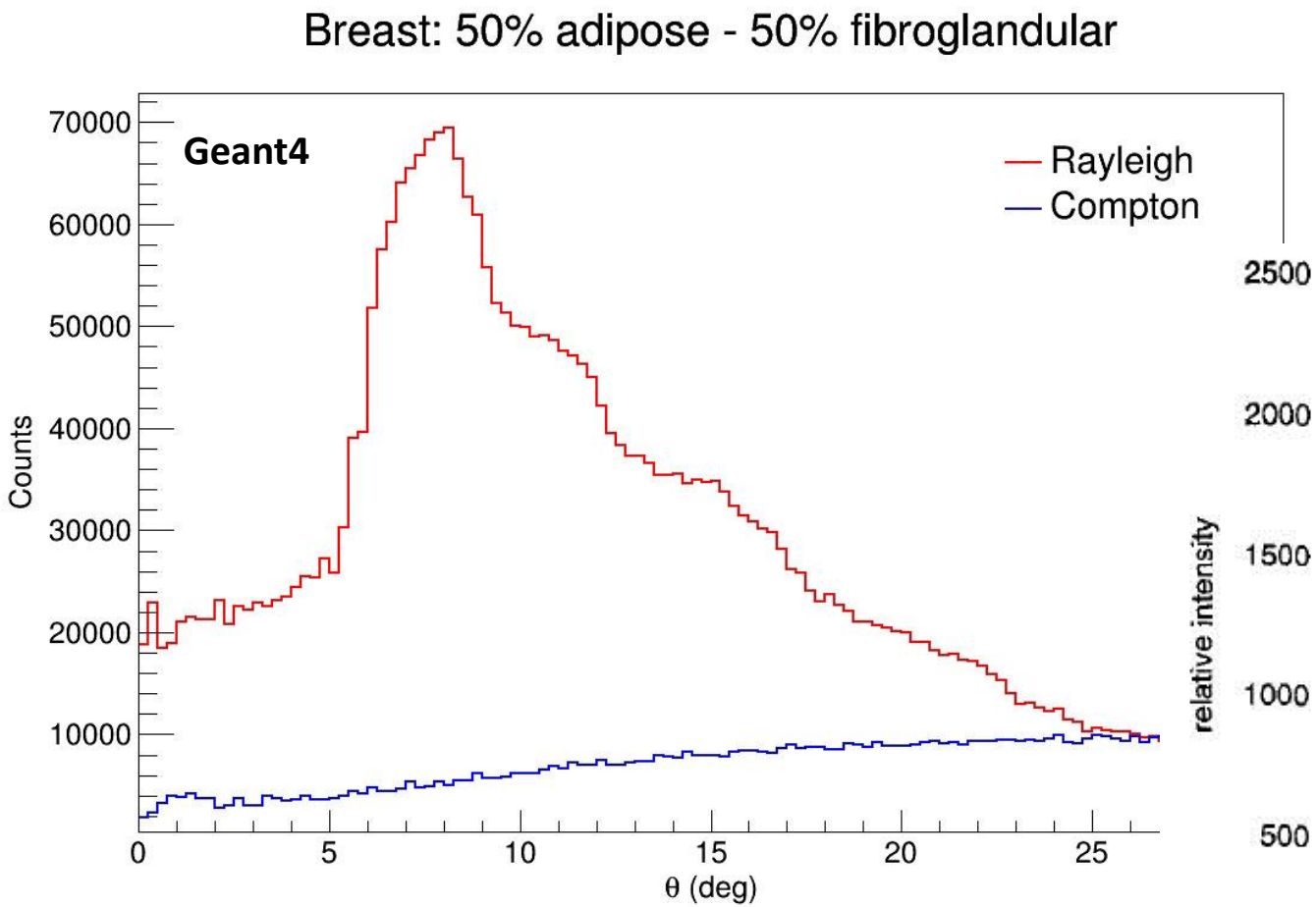


A **dedicated tool** has been developed in Geant4 to test the molecular interference implementation.

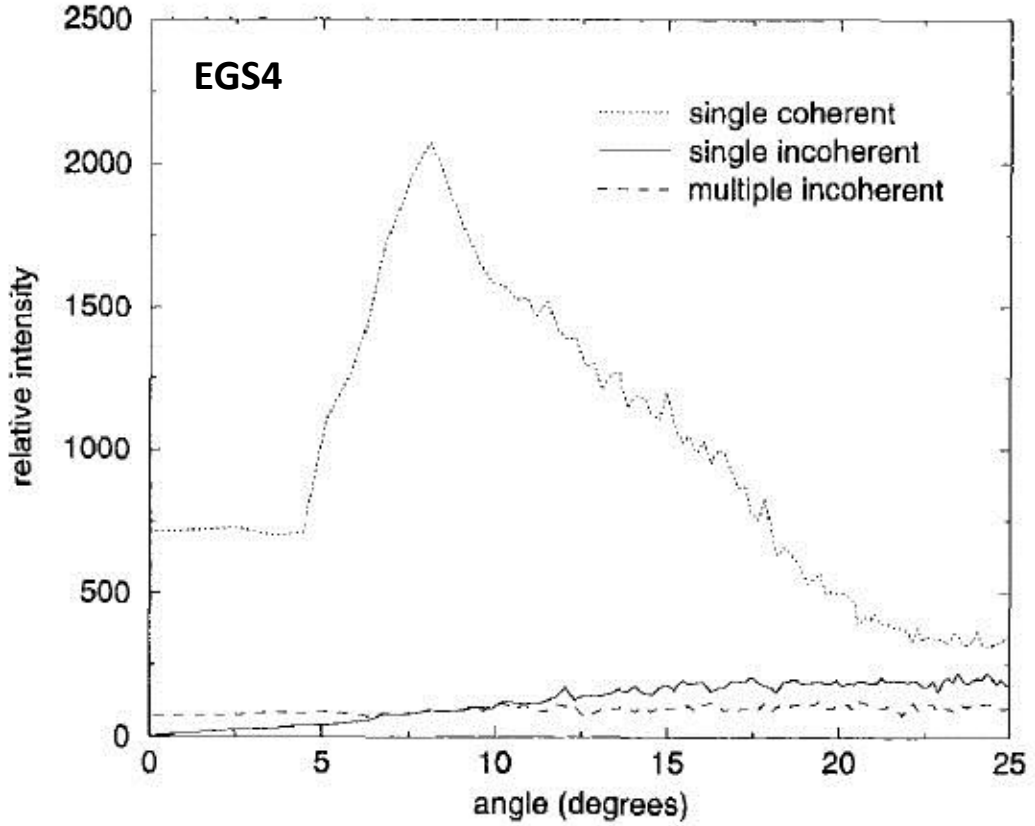
It involves a simple cylindrical phantom with a detail embedded. The phantom is irradiated with an X-ray beam and the scattered photons are scored.

- geometry management
- **material management** (the "**basis approach**" is foreseen and can be activated by codifying the material composition in its name, e. g., "*MedMat_0.25_0.36_0.13_0.36*")
- **various input beam** settable through macro
- **settable physics** and cuts
- scoring through SteppingAction and SensitiveDetector.

Validation

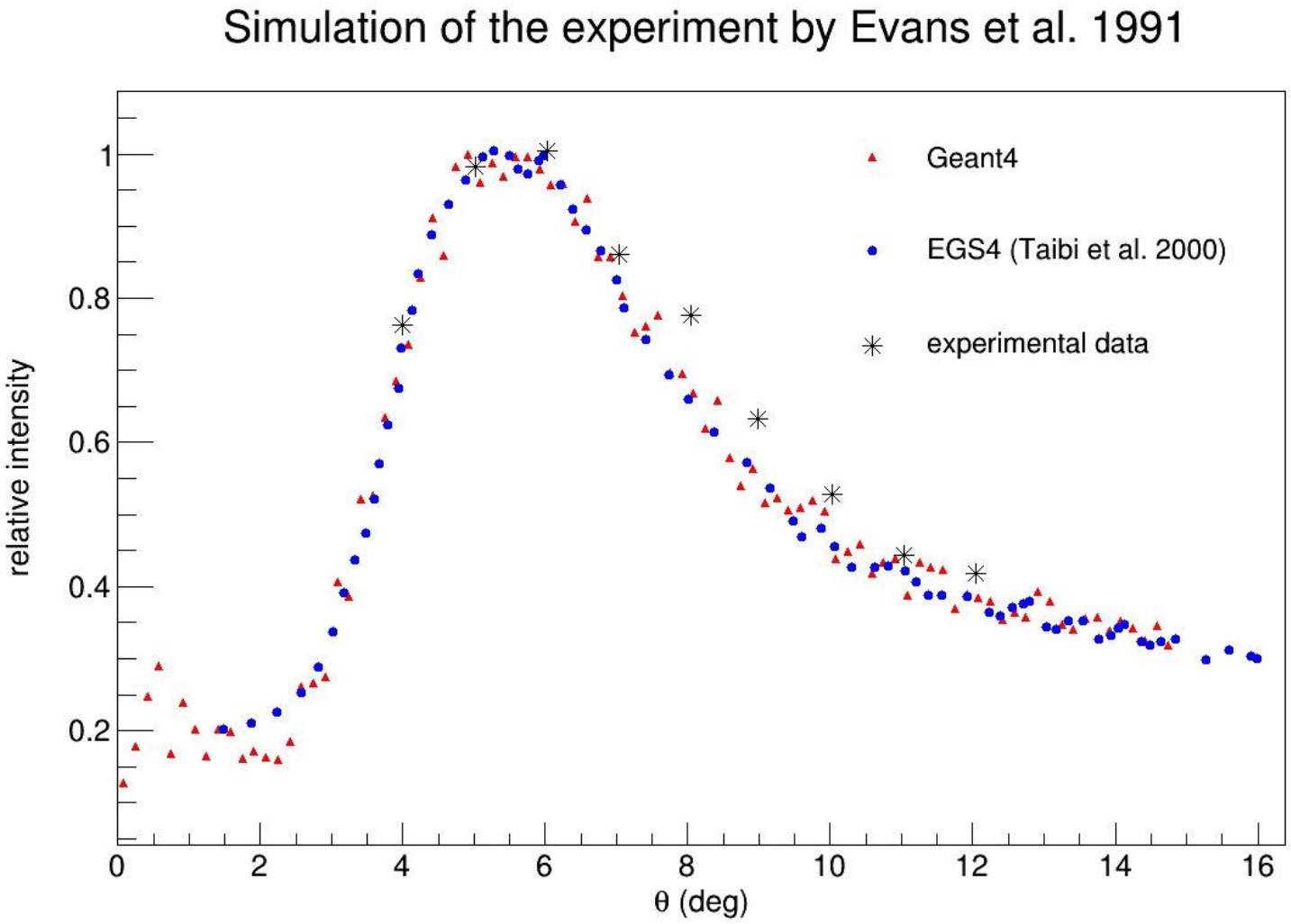


Taibi et al., IEEE trans. on nuclear science, vol 47 n. 4, 2000, 1581-1586.



Scatter profiles of **20 keV** photons impinging on a **5 cm-thick human breast sample.**

Validation

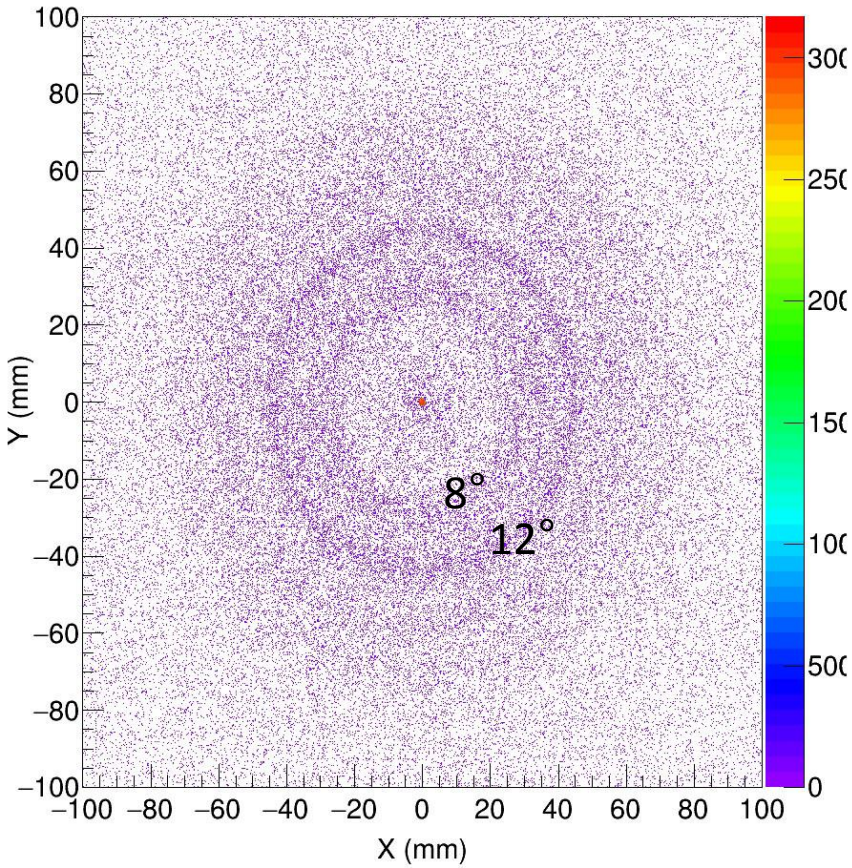


Scattering of **polychromatic X-rays** (60 kVp and filtration of 0.5 mm Cu) **from a 5 mm-thick carcinoma sample.**

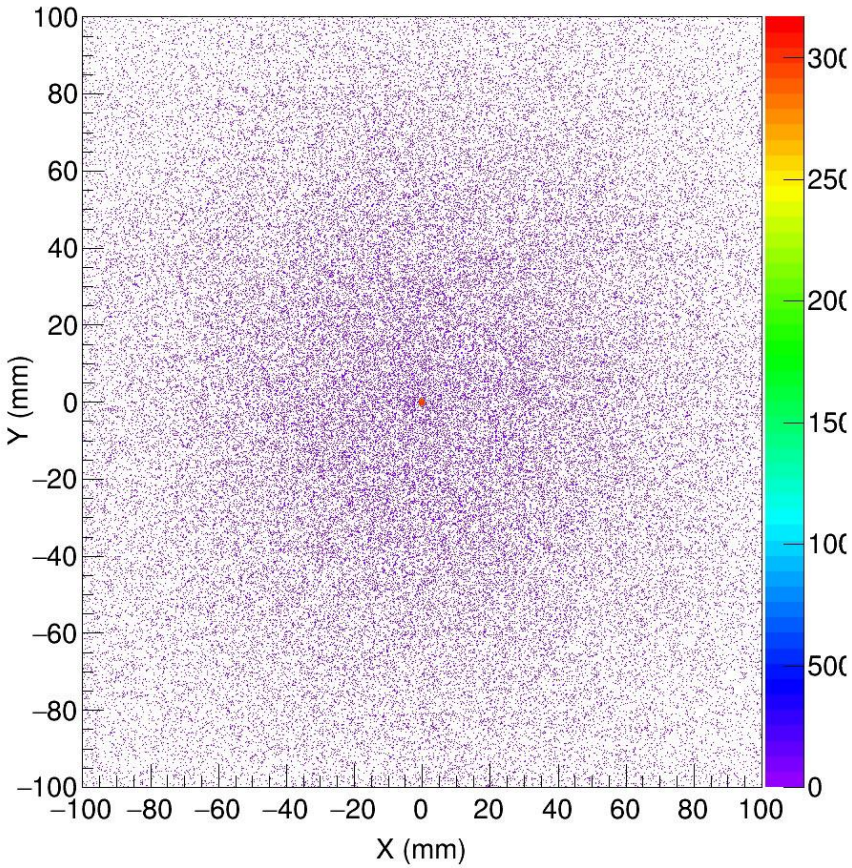
Simulations are in agreement with the experiment.

Application: rigorous simulation of SAXS

With Molecular Interference

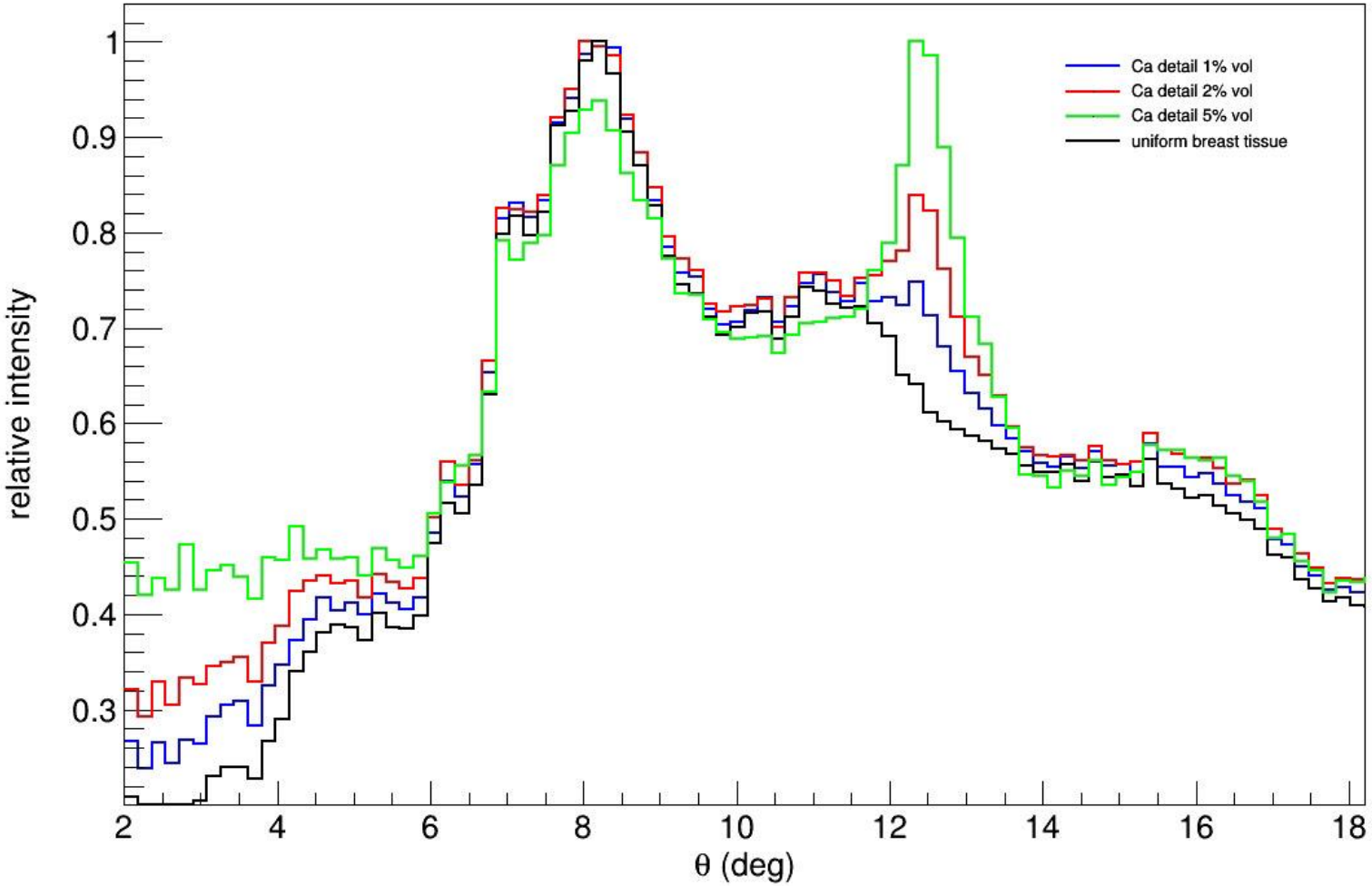


Without Molecular Interference



Scattering of a **20 keV pencil photon beam** impinging on a **5 cm-thick human breast** sample with a **1 mm-thick hydroxyapatite detail** embedded (simulating a calcification).

Application: identification of tissues



Scattering of a **20 keV** pencil photon beam incident on a **5 cm-thick human breast** sample with a **hydroxyapatite detail of various size** embedded (simulating a calcification).

Conclusions

• Molecular interference effect in coherent scattering has been implemented in Geant4 for a variety of materials.

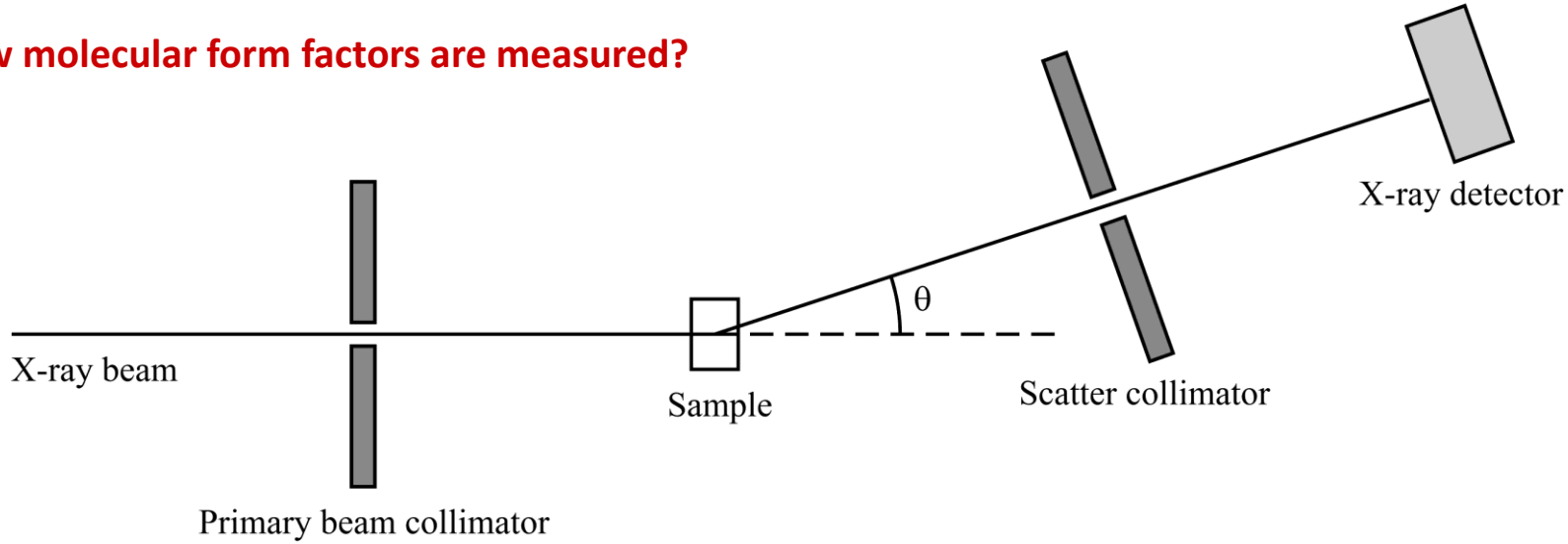
• The implementation has been validated comparing Geant4 simulations with previous results obtained through a different MC code and experimental data.

• The proposed updating will allow the user to simulate more rigorously scatter figures and SAXS experiments in Geant4.

Back-up slides

X-ray diffraction (XRD) experiments

How molecular form factors are measured?



ADXRD

- Scattering signal acquired as a function of θ
- Monochromatic X-ray beam
- Low photon flux
- Higher resolution achievable

EDXRD

- Scattering signal acquired at fixed angle θ
- Polychromatic X-ray beam
- Require a spectroscopic detector
- Faster

It is possible to combine these methods to improve the sensitivity *(see, for instance, Marticke et al., NIM A 867 (2017) 20-31)*

Simulation of coherent scattering events

```
291 //....oooOOOOOooo........oooOOOOOooo........oooOOOOOooo........oooOOOOOooo....
292 namespace { G4Mutex  PenelopeRayleighModelMutex = G4MUTEX_INITIALIZER; }
293 G4double G4PenelopeRayleighModel::ComputeCrossSectionPerAtom(const G4ParticleDefinition*,
294                                  G4double energy,
295                                  G4double Z,
296                                  G4double,
297                                  G4double,
298                                  G4double)
299 {
300   // Cross section of Rayleigh scattering in Penelope v2008 is calculated by the EPDL97
301   // tabulation, Cuellen et al. (1997), with non-relativistic form factors from Hubbel
302   // et al. J. Phys. Chem. Ref. Data 4 (1975) 471; Erratum ibid. 6 (1977) 615.
303
304    if (verboseLevel > 3)
305     G4cout << "Calling CrossSectionPerAtom() of G4PenelopeRayleighModel" << G4endl;
306
307    G4int iZ = (G4int) Z;
308
309    //Either Initialize() was not called, or we are in a slave and InitializeLocal() was
310    //not invoked
311    if (!logAtomicCrossSection)
312      {
313        //create a **thread-local** version of the table. Used only for G4EmCalculator and
314        //Unit Tests
315        fLocalTable = true;
316        logAtomicCrossSection = new std::map<G4int,G4PhysicsFreeVector*>;
317      }
318    //now it should be ok
319    if (!logAtomicCrossSection->count(iZ))
320      {
321        //If we are here, it means that Initialize() was inkoved, but the MaterialTable was
322        //not filled up. This can happen in a UnitTest or via G4EmCalculator
323        if (verboseLevel > 0)
324     {
325        //Issue a G4Exception (warning) only in verbose mode
326        G4ExceptionDescription ed;
327        ed << "Unable to retrieve the cross section table for Z=" << iZ << G4endl;
328        ed << "This can happen only in Unit Tests or via G4EmCalculator" << G4endl;
329        G4Exception("G4PenelopeRayleighModel::ComputeCrossSectionPerAtom()",
330                "em2040",JustWarning,ed);
331     }
332        //protect file reading via autolock
```

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Simulation of coherent scattering events

```
664 //....oooO0000ooo........oooO0000ooo........oooO0000ooo........oooO0000ooo....
665
666 void G4PenelopeRayleighModel::SampleSecondaries(std::vector<G4DynamicParticle*>* ,
667                         const G4MaterialCutsCouple* couple,
668                         const G4DynamicParticle* aDynamicGamma,
669                         G4double,
670                         G4double)
671 {
672   // Sampling of the Rayleigh final state (namely, scattering angle of the photon)
673   // from the Penelope2008 model. The scattering angle is sampled from the atomic
674   // cross section dOmega/d(cosTheta) from Born ("Atomic Phyisics", 1969), disregarding
675   // anomalous scattering effects. The Form Factor F(Q) function which appears in the
676   // analytical cross section is retrieved via the method GetFSquared(); atomic data
677   // are tabulated for F(Q). Form factor for compounds is calculated according to
678   // the additivity rule. The sampling from the F(Q) is made via a Rational Inverse
679   // Transform with Aliasing (RITA) algorithm; RITA parameters are calculated once
680   // for each material and managed by G4PenelopeSamplingData objects.
681   // The sampling algorithm (rejection method) has efficiency 67% at low energy, and
682   // increases with energy. For E=100 keV the efficiency is 100% and 86% for
683   // hydrogen and uranium, respectively.
684
685   if (verboseLevel > 3)
686     G4cout << "Calling SamplingSecondaries() of G4PenelopeRayleighModel" << G4endl;
687
688   G4double photonEnergy0 = aDynamicGamma->GetKineticEnergy();
689
690   if (photonEnergy0 <= fIntrinsicLowEnergyLimit)
691     {
692       fParticleChange->ProposeTrackStatus(fStopAndKill);
693       fParticleChange->SetProposedKineticEnergy(0.);
694       fParticleChange->ProposeLocalEnergyDeposit(photonEnergy0);
695       return ;
696     }
697
698   G4ParticleMomentum photonDirection0 = aDynamicGamma->GetMomentumDirection();
699
700   const G4Material* theMat = couple->GetMaterial();
701
702
703   //1) Verify if tables are ready
704   //Either Initialize() was not called, or we are in a slave and InitializeLocal() was
705   //not invoked
706   if (!pMaxTable || !samplingTable || !logAtomicCrossSection || !atomicFormFactor ||
707       !logFormFactorTable)
708     {
```

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Implementation

DetectorConstruction.cc × G4PenelopeRayleighModel.cc ×

```
179         matname = "BoneMatrix";
180     } else {
181         matname = "BoneMatrix_noint";
182     }
183     BoneMatrix = new G4Material(matname, d_BoneMatrix, nel);
184     BoneMatrix->AddElement(elH, 0.0344);
185     BoneMatrix->AddElement(elC, 0.7140);
186     BoneMatrix->AddElement(elN, 0.1827);
187     BoneMatrix->AddElement(elO, 0.0689);
188
189     //Mineral (Hydroxyapatite)
190     G4double d_Mineral = 2.74*g/cm3;
191     nel = 4;
192     if (IWantMI) {
193         matname = "Mineral";
194     } else {
195         matname = "Mineral_noint";
196     }
197     Mineral = new G4Material(matname, d_Mineral, nel);
198     Mineral->AddElement(elH, 0.002);
199     Mineral->AddElement(elO, 0.414);
200     Mineral->AddElement(elP, 0.185);
201     Mineral->AddElement(elCa, 0.399);
202
203     //Medical Material
204     G4double comp[] = {Comp0, Comp1, Comp2, Comp3}; //enter the material composition
205     G4double d_MedMat = d_Fat*d_Water*d_BoneMatrix*d_Mineral/(comp[0]*d_Fat*d_BoneMatrix*d_Mineral+comp[1]*d_Water*d_BoneMatrix*d_Mineral+comp
    [2]*d_Fat*d_Water*d_Mineral+comp[3]*d_Fat*d_Water*d_BoneMatrix);
206     G4cout << "### MedMat density: " << d_MedMat/(g/cm3) << " ###" << G4endl;
207     G4int n_MedMat = 0;
208     for (size_t i=0; i<4; i++) {
209         if (comp[i]>0) n_MedMat++;
210         if (comp[i]<0 || comp[i]>1) {
211             G4String excep = "Error in Medical Material composition: comp[i]<0 or comp[i]>1";
212             G4Exception("DetectorConstuction::DefineMaterials()",
213             "em0001",FatalException,excep);
214             return;
215         }
216     }
217     std::stringstream ss0,ss1,ss2,ss3;
218     ss0 << comp[0];
219     ss1 << comp[1];
220     ss2 << comp[2];
221     ss3 << comp[3];
```

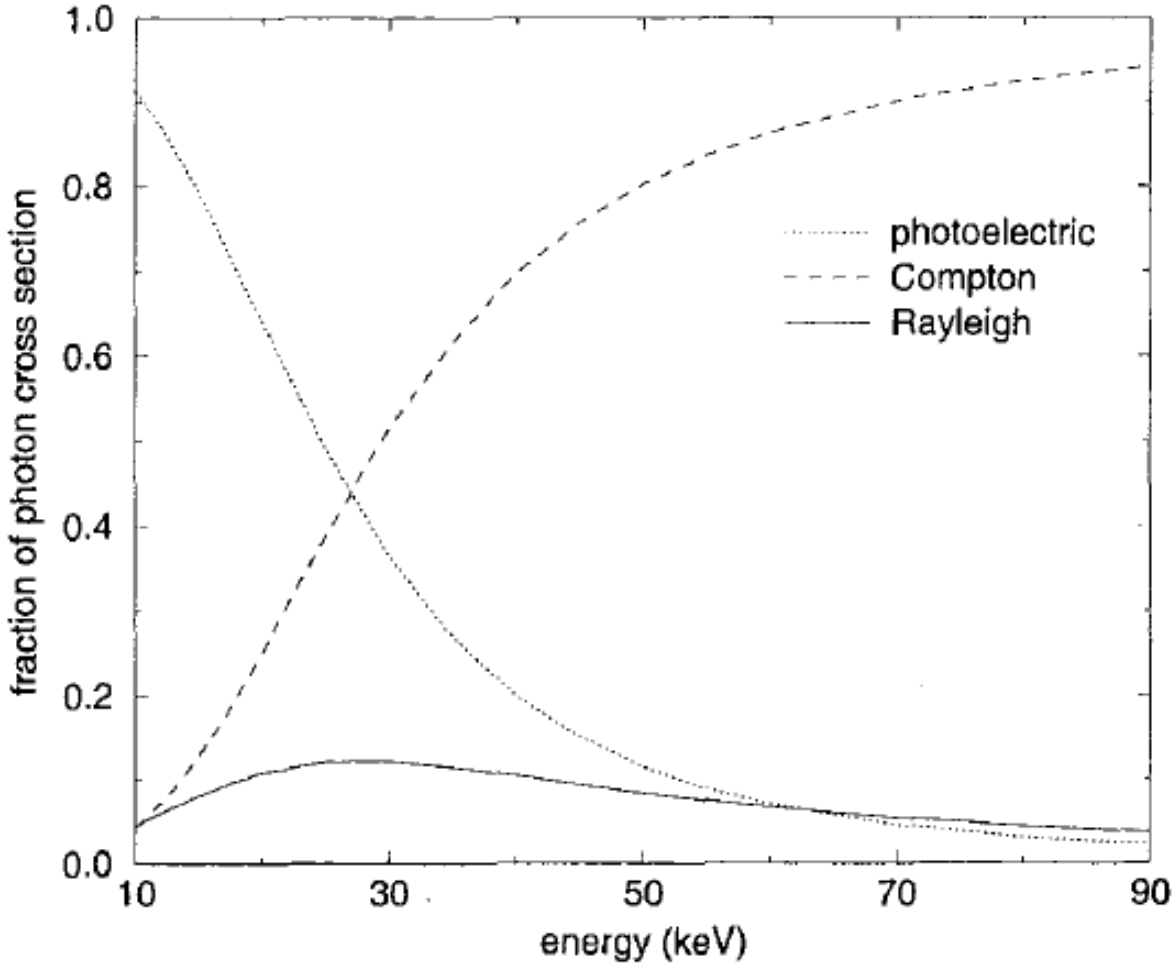
C++ ▾ Tab Width: 4 ▾

Implementation

DetectorConstruction.cc × G4PenelopeRayleighM... Copy the selection

```
438  G4PhysicsFreeVector* graymatterFF = MolInterferenceData->find(24)->second;
439
440  G4String matname = material->GetName();
441
442  //medical material: composition of fat, water, bonematrix, mineral
443  if (matname.find("MedMat") != std::string::npos) {
444    G4cout << "MIFF: MedMat" << G4endl;
445
446    //get the material composition from its name
447    G4int ki, kf=6, ktot=19;
448    G4double comp[4];
449    G4String compstring = matname.substr(kf+1, ktot);
450    for (size_t j=0; j<4; j++) {
451        ki = kf+1;
452        kf = ki+4;
453        compstring = matname.substr(ki, 4);
454        comp[j] = atof(compstring.c_str());
455        G4cout << "MedMat comp[" << j+1 << "]: "  << comp[j] << G4endl;
456    }
457
458    //get and combine the molecular form factors with interference effect
459    for (size_t k=0;k<logQSquareGrid.size();k++) {
460        G4double ff2 = 0;
461        G4double ffat = (*fatFF)[k];
462        G4double fwater = (*waterFF)[k];
463        G4double fbonematrix = (*bonematrixFF)[k];
464        G4double fmineral = (*mineralFF)[k];
465        ff2 = comp[0]*ffat*ffat+comp[1]*fwater*fwater+comp[2]*fbonematrix*fbonematrix+comp[3]*fmineral*fmineral;
466        if (ff2) theFFVec->PutValue(k,logQSquareGrid[k],std::log(ff2));
467    }
468  }
469
470  //other materials with interference function
471  else if (matname == "PMMA") {
472    G4cout << "MIFF: PMMA" << G4endl;
473    for (size_t k=0;k<logQSquareGrid.size();k++) {
474        G4double ff2 = 0;
475        G4double f = (*PMMAFF)[k];
476        ff2 = f*f;
477        if (ff2) theFFVec->PutValue(k,logQSquareGrid[k],std::log(ff2));
478    }
479  } else if (matname == "Adipose") {
480    G4cout << "MIFF: Adipose" << G4endl;
```

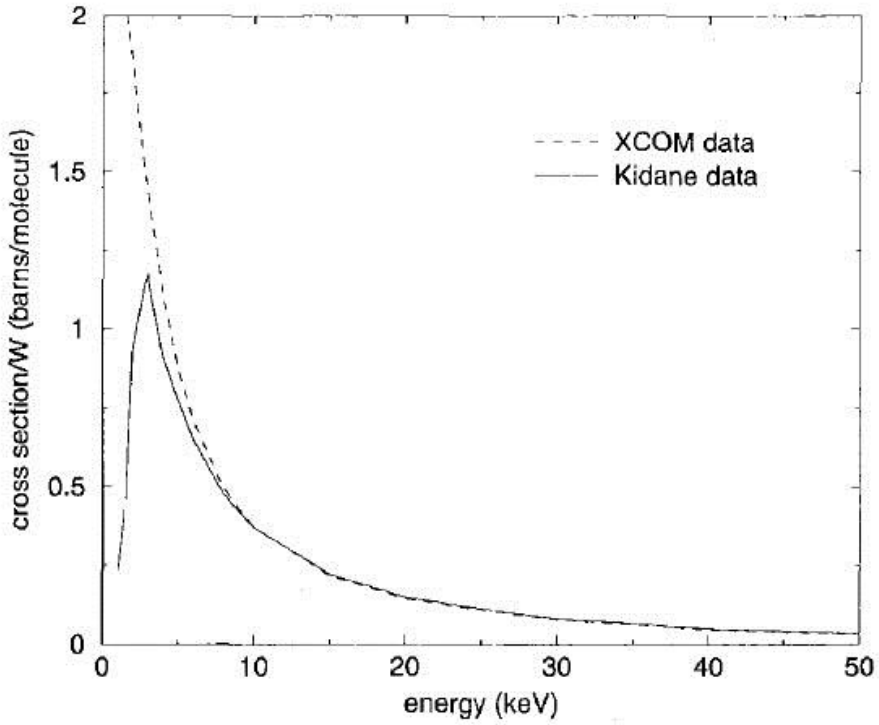
Theoretical background: Coherent Scattering



The fraction of coherent scattering interactions is about 10% for materials and energies of medical interest but, due to MI, coherent radiation is not forward peaked and is distinguishable from primary radiation.

MI effect implementation in Geant4

Since coherent scattering total cross-section for compounds is managed by a separate class ad it remains approximately the same with and without MI for energies of medical interest (see the figure), **we used modified form factors only for the sampling of the photon angular deflection**.



*Comparison of calculated coherent cross-section for **carcinoma** with and without molecular interference (Taibi et al., IEEE trans. on nuclear science, vol 47 n. 4, 2000, 1581-1586).*

Simulation of coherent scattering events (Penelope algorithm)

First, the occurrence of a coh. scatt. event is determined from σ_{Ra}, then the angular deflection is sampled

$$P_{Ra}(\cos\theta) = \frac{1+\cos^2\theta}{2} F^2(q) \qquad 0 \le q \le q_{\max} = 2E/c = 2m_e c\kappa$$

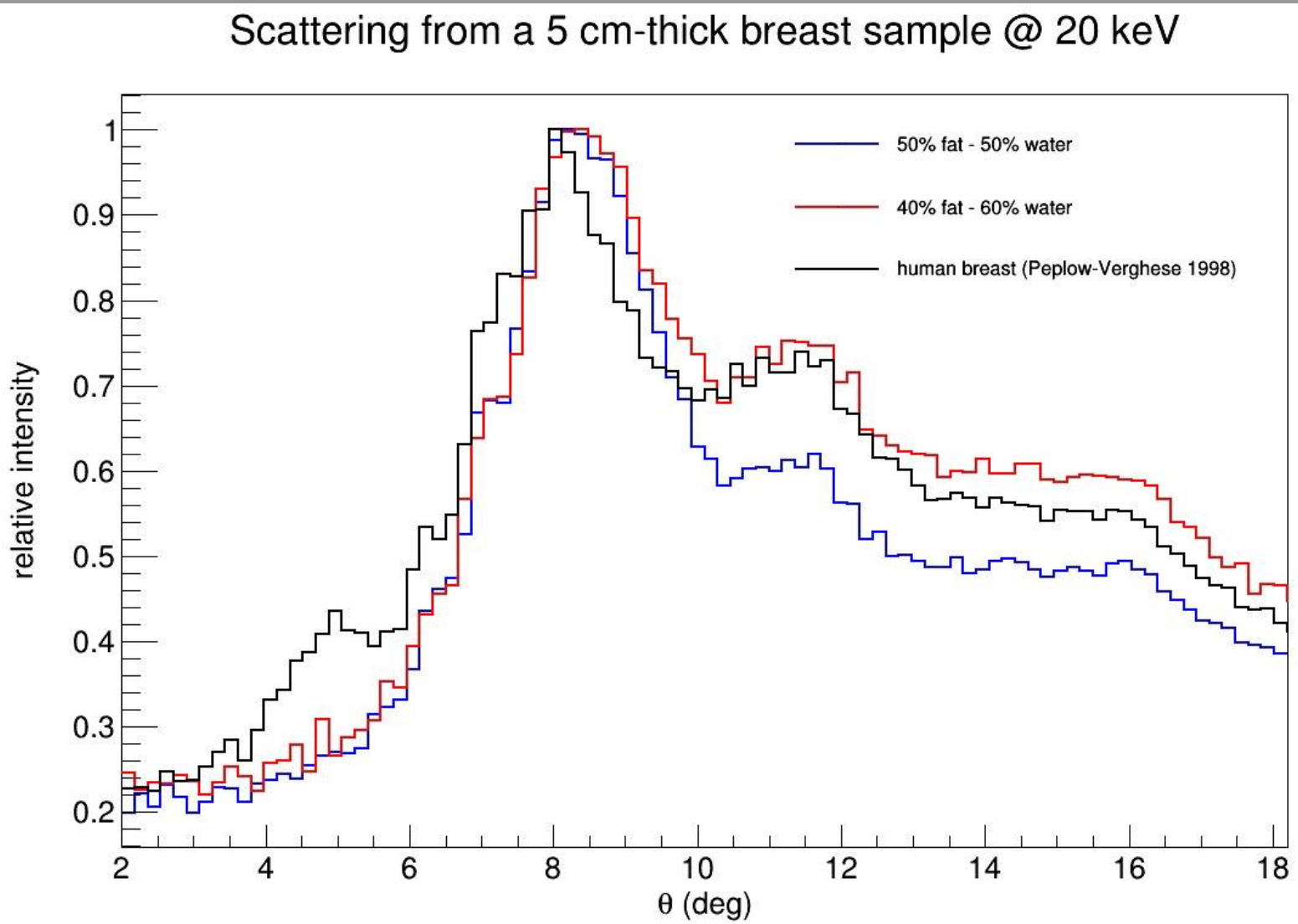
$$P_{Ra}(\cos\theta) = g(\cos\theta)\pi(q^2) \qquad g(\cos\theta) = \frac{1+\cos^2\theta}{2} \qquad \pi(q^2) = F^2(q)$$

rejection method

1. Using the **RITA algorithm**, sample a random value of q^2 from the distribution $\pi(q^2)$, restricted to the interval $[0, q_{max}^2]$.
2. Set $\cos\theta = 1 - 1/2 * q^2/k^2$ ($k = E/m_e c^2$). *(it comes from the definition of $q = 2E/c[\sin(\vartheta/2)] = (E/c[2(1-\cos\vartheta)]^{1/2})$*
3. Generate a new random number ξ (uniformly distributed in the interval [0,1]).
4. If $\xi > g(\cos\theta)$, go to step 3. *(note that **g is a valid rejection function** since $0 < g \le 1$)*
5. Deliver $\cos\theta$.

Sampling efficiency higher than 66%

Application: identification of tissues



Application: identification of cancer signatures