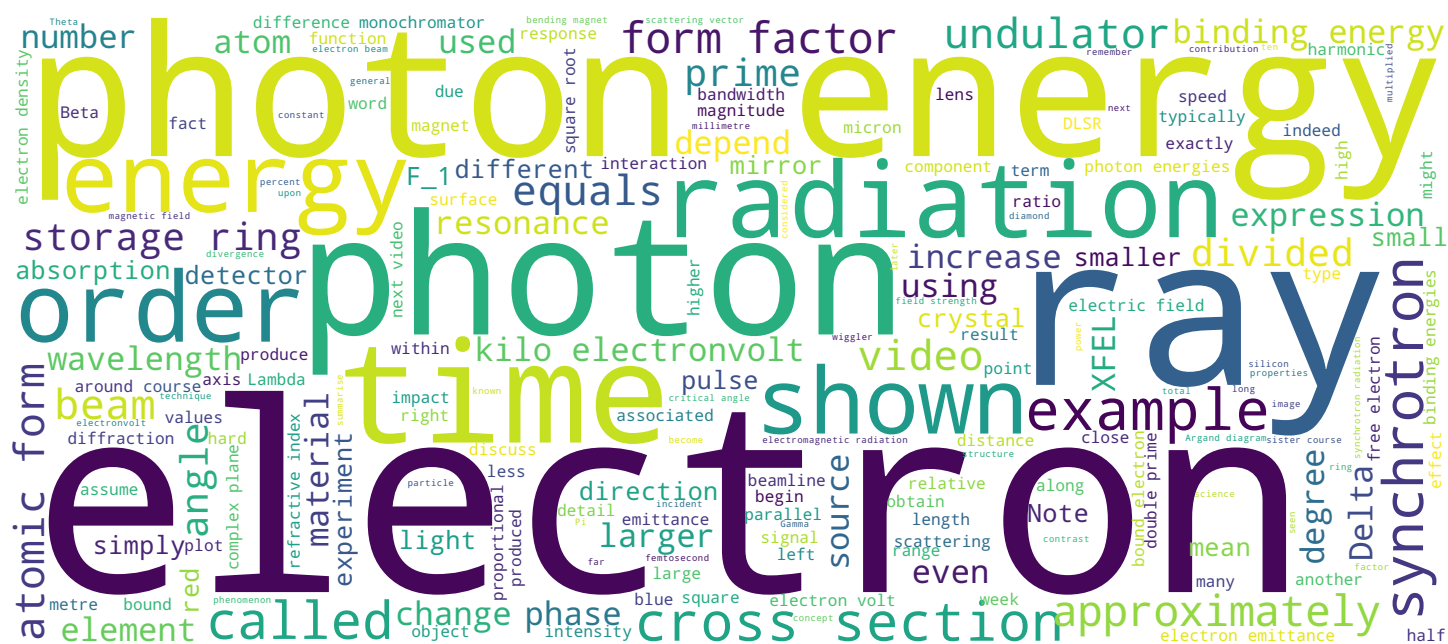


Synchrotrons and x-ray free-electron lasers

Techniques and applications

■ École polytechnique fédérale de Lausanne



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Video



Contents and objectives of this video



- Why correction terms to f are needed
- The impact of electrons being bound

In this video, we consider how scattering of radiation by the electrons of an atom is affected by the fact that the electrons are bound. This leads to the addition of correction terms that change with photon energy, that is, they are dispersive.

Notes

Summary



0m 05s

f^0 and beyond

$$f^0(\sin \theta / \lambda) = \sum_{i=1}^4 a_i \exp(-b_i \sin^2 \theta / \lambda^2) + c$$

Our discussion thus far has ignored the phenomenon of absorption of radiation. We've assumed that all the electrons that make up the electron cloud can respond instantaneously and with no damping to the incident, oscillating electromagnetic wave. In reality, electrons are bound to atoms and assumed discrete energy levels determined by quantum mechanics, or in the case of valence electrons for atoms in condensed matter, energy bands. Thus, the electrons have defined binding energies. The response of bound electrons depends on this binding energy and the energy of the incident photon. We will consider three cases: one for which the photon energy is much smaller than the binding energy of the electron, one where it is much larger, and finally, the case of the photon energy being exactly equal to the binding energy, a condition known as resonance. In the last video, we described the atomic form factor of the elements as a function of the scattering vector Q , or equivalently sine feature upon Λ , using the expression shown here. Note that F has a superscript zero, which we highlight here in red.

Notes

Summary



0m 23s

f^0 and beyond

$$f^0(\sin \theta / \lambda) = \sum_{i=1}^4 a_i \exp(-b_i \sin^2 \theta / \lambda^2) + c$$

This denotes that this expression is the simplest description of F , which assumes the electrons are unhindered by their response to the incoming x-rays, by the fact that they are bound to atomic nuclei. The photon energy is much larger than the electrons binding energy, this is actually a valid approximation.

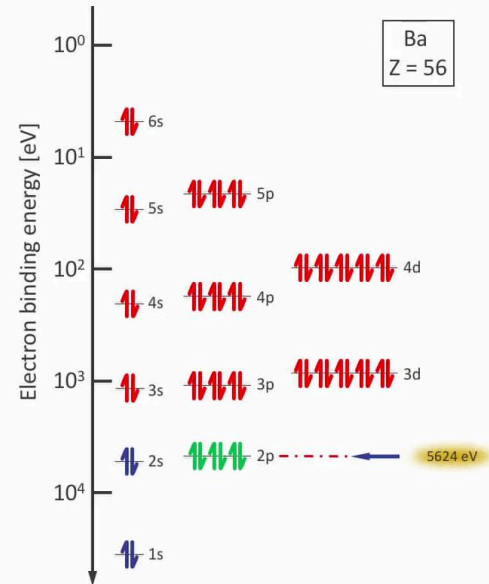
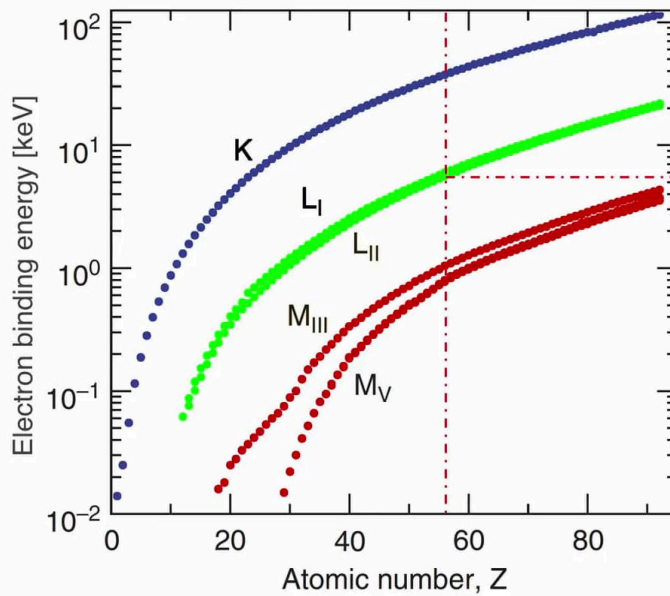
Notes

Summary

1m 52s



Electron binding energies of the elements



As I'm sure most of you already know, electrons in atoms assume well-defined energies enforced by the laws of quantum mechanics. The most strongly bound are the core level 1s or K electrons, followed by the electrons from the n equals 2 shell, alternatively called L electrons, and so on and so forth. On the left is a plot of the binding energy of the elements up to uranium at z equals 92. On the right is shown the energy level scheme of the electronic configuration of the element barium, which as a neutral atom, contains 56 electrons. If we irradiate barium with photons resonant with, for example, an L electron, this will have a strong absorption cross-section and is said to be at resonance. Electrons with higher binding energies, shown here in blue, cannot absorb this photon and remain at their energy level. Those with lower binding energies can absorb the photon, but with a lower cross-section than that of the resonant electron, which can be promoted to an unbound excited state.

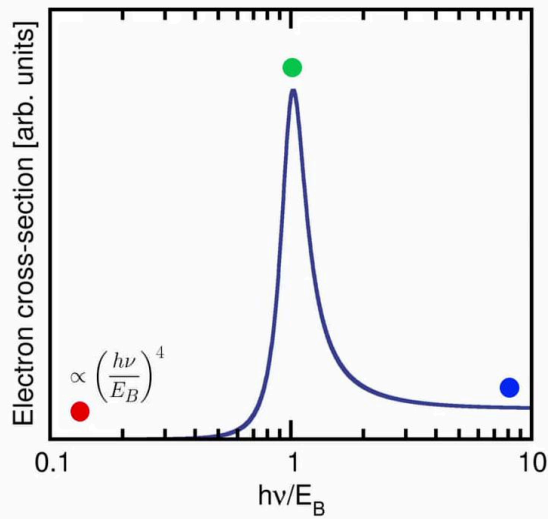
Notes

Summary

2m 15s



Bound electrons' response to x-rays



- $h\nu \ll E_B$
 - Response strongly suppressed ~ 0
 - "Rayleigh scattering"
- $h\nu \gg E_B$
 - Electron quasi "free"
 - $\phi = \pi$ - scattered radiation out of phase with incident beam
- $h\nu \simeq E_B$
 - Resonance
 - Enhanced response
 - $\phi = \pi/2$: dissipation (absorption)

How exactly does a bound electron respond to instant electromagnetic radiation? We can model this as a damped oscillator responding to an oscillatory driving force. The system has a natural oscillation frequency given by ω_0 , which is equal to E_b divided by \hbar . If the driving frequency ω is much smaller than ω_0 , the response amplitude and associated cross-section are strongly suppressed by the electron being bound. The cross-section drops off as the approximately fourth power of the ratio of $h\nu$ to E_b and it's called Rayleigh scattering.

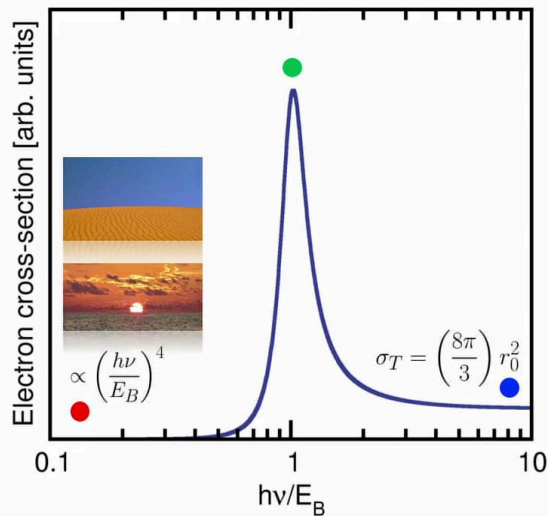
Notes

Summary



3m 33s

Bound electrons' response to x-rays



- $h\nu \ll E_B$
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- $h\nu \gg E_B$
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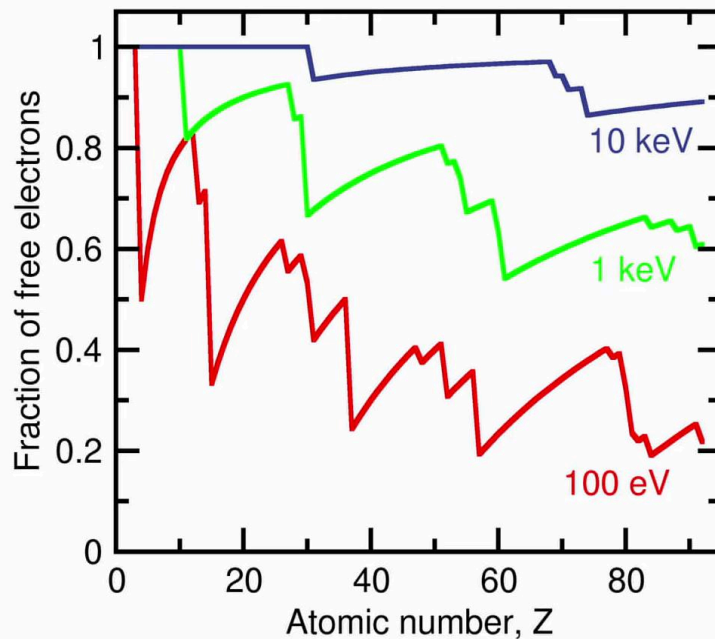
This is the reason, incidentally, why the sky is blue and sunrises and sunsets are red. In the former case, the blue end of the broadband radiation from the sun is scattered more efficiently than the red by microscopic fluctuations in the air density. Sunrises and sunsets are concentrated around the rising or setting sun. In these instances, the blue part of the solar spectrum is more efficiently scattered away from the observer who is looking directly towards the sun, leaving behind the characteristic red hue. Anyway, for hard x-rays, however, a good fraction of the electrons will have binding energies that are much smaller than the photon energy. Thus, for photon energies far above resonance, the electrons respond to excitation by the photons essentially as if they were free, and the cross-section approaches that of the free electron, the Thomson cross-section we encountered earlier in these weeks videos. The re-emitted radiation has a phase of π , or 180 degrees, relative to the instant beam due to the electron response being exactly out of phase with the instant radiation. At resonance, the electrons cross-section is enhanced and the phase of the re-emitted radiation is 90 degrees, or π upon 2, which is associated with absorption, as we will discuss shortly.

Notes

Summary



Atomic response to x-rays



Shown here is a plot of the fraction of electrons in the naturally occurring elements that have binding energies smaller than 100 electronvolts in red, and one kilo-electronvolt shown in green, and 10 kilo-electronvolt photons shown in blue, and can thus be considered to be quasi free with regards to their response to and cross-sections for interactions with those photons. We see that for the soft x-rays at 100 electronvolts, a good fraction of the electrons remain bound. Things change rapidly with photon energy, however. Even at one kilo-electronvolt, at most about 45 percent of the electrons are bound, for the case of the element promethium at z equals 61, which incidentally, is not the most relevant of elements, there being about half a kilo of it in the entire Earth's crust, it being a radioactive decay product. At 10 kilo-electronvolts, the photons are approximately 35 times more energetic than the most strongly bound electron in carbon, while even in uranium, which contains 92 electrons, only the 10 K and L electrons are more tightly bound than 10 kilo-electronvolts.

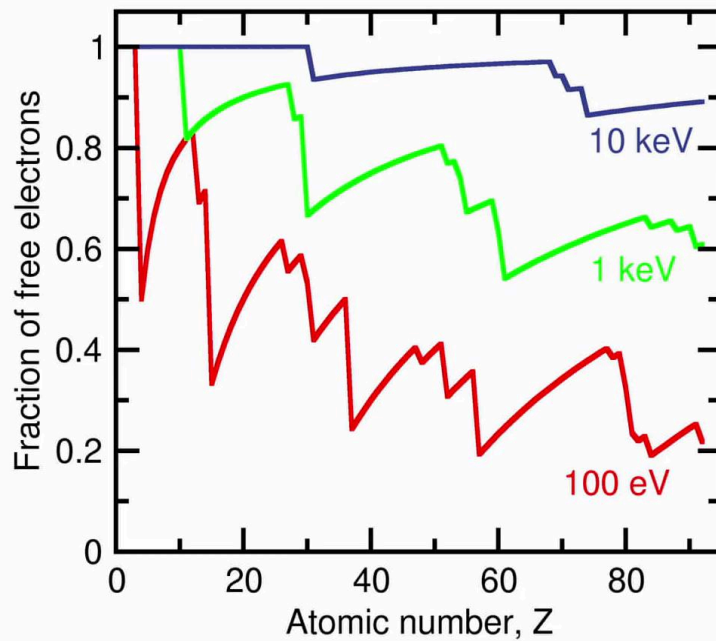
Notes

Summary



5m 50s

Atomic response to x-rays



Take-home message:

Most electrons in atoms can be considered to be quasi "free" for $h\nu > \text{keV}$

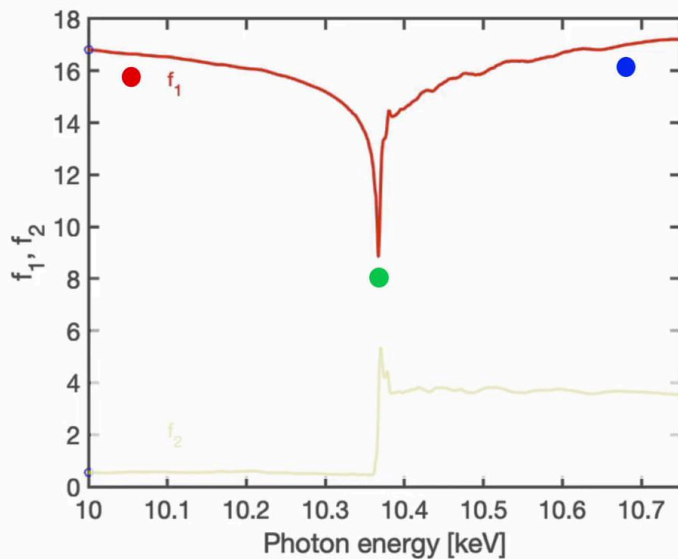
So the take-home message is, therefore, that for x-rays with photon energies of the order of a kilo-electronvolt or higher, most electrons and all the elements can be considered to be quasi free with regards to their response to and cross-sections for interactions with those photons. Nonetheless, the impact of resonance is crucial for many phenomena, which we will discuss shortly. First, we need to formalise the impact of resonances on the atomic form factor F .

Notes

Summary



Correction terms to f: f'



- $h\nu < E_B$
 - Reduced response from those electrons that are bound \Rightarrow small reduction in scattering factor
 - Add **negative** component f'
 - f' a function of $h\nu$
- $h\nu \gg E_B$
 - Electron quasi "free"
 - $f' \Rightarrow 0$
- $h\nu \approx E_B$
 - Resonance
 - Enhanced response \Rightarrow maximal $|f'|$

$$f_1(Q, \hbar\omega) = f^0(Q) + f'(\hbar\omega)$$

$$f'(\hbar\omega) < 0$$

Our expression for the atomic form factor so far needs to be modified by two extra terms that depend on the photon energy called F' and F'' . Because of their dependence on photon energy, these are referred to as dispersive terms. We begin with F' . This is a negative correction to the atomic form factor, which describes the fact that the bound electrons have a damped resonance. F_1 is the sum of the free electron atomic form factor F_0 , which depends only on the scattering vector Q and F' , which only depends on the instant photon energy. Because F' is negative, F_1 is smaller than F_0 , as shown in the plot here in red for the element gallium. It assumes its smallest value at resonance, where the photon energy is close to the electron binding energy.

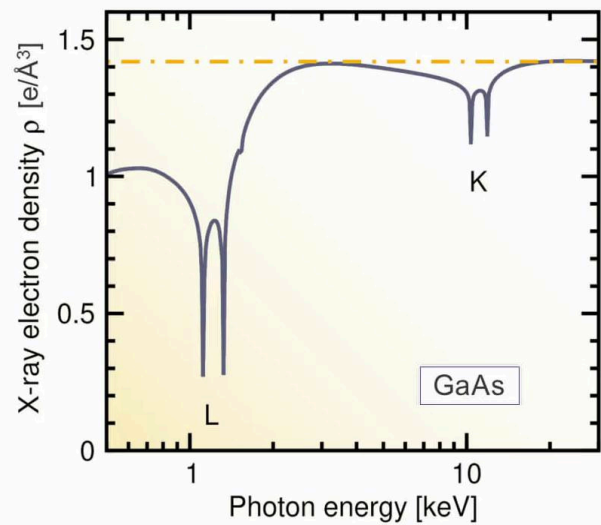
Notes

Summary



f' accounts for damped oscillation amplitude

- Effect of f' is to make the atom “appear” from the perspective of the x-rays to have fewer electrons ($< Z$) than far above an absorption edge
- ρ as “seen” by x-rays decreases
- Scattering strength decreases near absorption edges



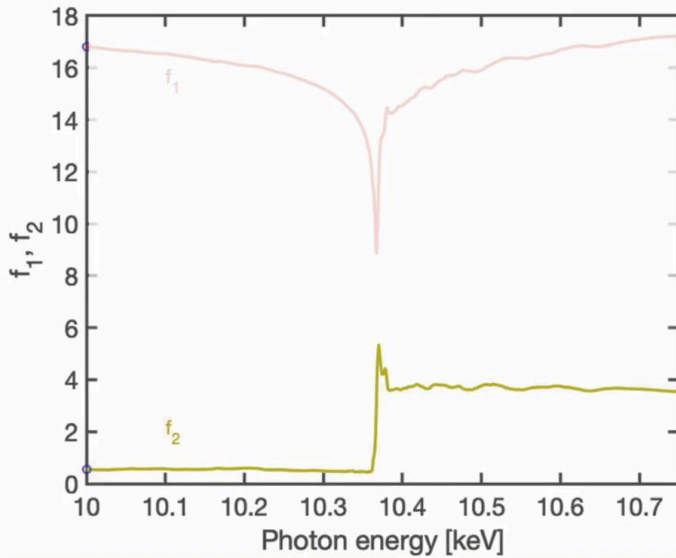
F prime therefore accounts for the damping in the electrons oscillation amplitude due to it being bound to the atom. The effect of F prime is therefore to make the atom appear to have a lower electron density than it actually has, at least from the perspective of the x-rays. One can calculate the apparent electron density as seen by the x-rays, by performing x-ray reflectivity measurements. We'll look at this later in this week's videos. We see here an example for the compound, semiconductor gallium arsenide. The atomic numbers of gallium and arsenic are 31 and 33, hence their absorption edges lie close to one another, resulting in the doublets of dips seen here for their K and L-edges. Only in between these absorption edges, and again at photon energy significantly higher than the K-edge, does the electron density, as perceived by the x-rays, agree well with the known physical values of approximately 1.4 electrons per cubic angstrom, shown as the dot-dashed line in gold.

Notes

Summary



Correction terms to f: f''



- $h\nu \approx E_B$
 - Resonance
 - Enhanced response \Rightarrow maximal $|f'|$
 - Phase shift @ resonance = $\pi/2$
 - Express as imaginary component if''
 - f' a function of $h\nu$
 - Results in energy dissipation (absorption)

$$f_2 = f'' = \frac{\sigma_a}{2\lambda r_0}$$

Close to the binding energy of an electron, not only is F_1 enhanced, but also a second term becomes important called F_2 . Because the resonant electron has a phase of 90 degrees to the instant radiation, this term is multiplied by i , the square root of minus one in order that the vector description of the atomic form factor when plotted in the complex plane in an Argand diagram, it lies at 90 degrees to F_1 . F_2 , also called F_2' , is therefore the imaginary component of the total form factor and F_1 is the real component. F_2 results in energy dissipation due to photo absorption and is equal to the absorption cross-section divided by $2\lambda r_0$. Why we need to multiply F_2 by i will become immediately apparent. Note also that both F_1 and F_2 have fine details as we see here in this graph consisting of small oscillations. These are not experimental artefacts, but are in fact important features from which detailed information can be drawn in methods such as [inaudible 00:11:25] covered in the sister course.

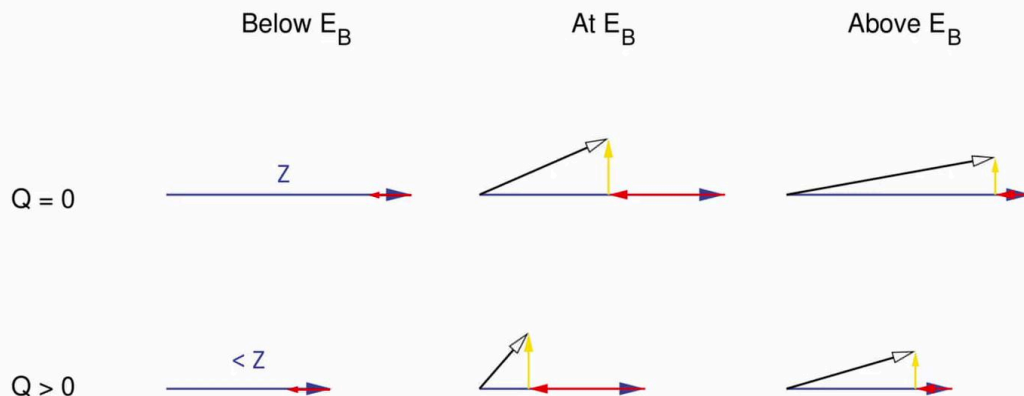
Notes

Summary

10m 06s



Summary of correction terms f' and f''



$$f(Q, \hbar \omega) = \underbrace{f^0(Q)}_{f_1(Q, \hbar \omega)} + \underbrace{f'(\hbar \omega) + if''(\hbar \omega)}_{f_2(\hbar \omega)}$$

See also description of vector addition in Argand diagrams in the supplementary text "Argand diagram summary"

We summarise these two additional and energy dependent terms here. F_0 shown as the blue arrow depends only on the scattering vector. It is equal to z for q equals 0, as we have already discussed, and assume smaller values for non-zero Q values. At resonance, where the photon energy equals the binding energy of an electron in the atom, F_0 is reduced by F prime, shown as the red arrow, and assumes the value F_1 . At right angles to this, F double prime or F_2 is added vectorially. The 90 degree angle is the phased difference to F_1 given implicitly by the factor i in the complex plane. As the energy increases further, the damping term F prime drops off again, as does F_2 , though more slowly. For those of you unfamiliar with vector addition in the complex plane, look also at the description of vector addition using Argand diagrams in the supplementary text, Argand diagram summary.

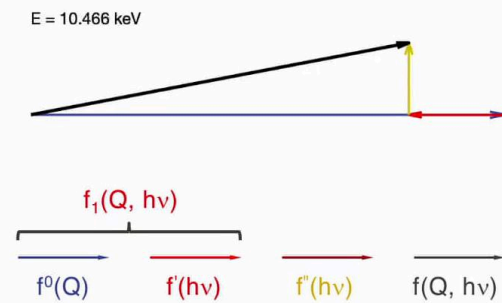
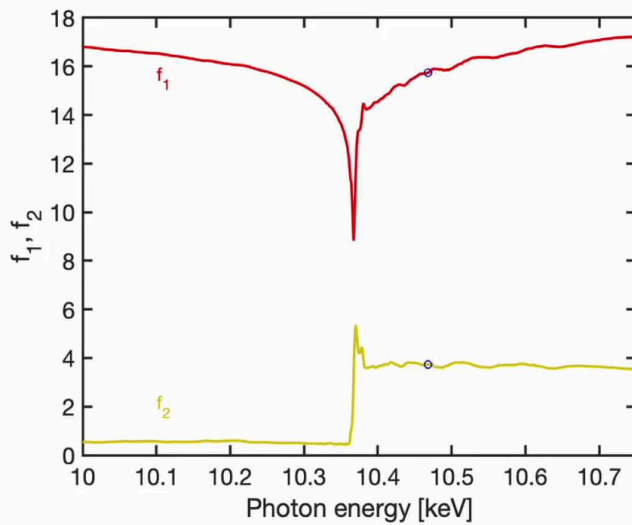
Notes

Summary



11m 28s

Change in f for Ga @ K-edge and $Q = 4.45 \text{ \AA}^{-1}$



To summarise this video, let's look at the real case of the change in the total atomic scattering factor close to the K-edge absorption, that is the excitation of a 1s electron in the element gallium. The values of F_1 and F_2 are tracked as a function of the instant photon energy by the small blue circles in the animated plot on the left. In parallel, the change in each component and the resulting atomic form factor are shown by the vector addition on the right. So let's go. To begin with, not a great deal happens other than F_1 slowly getting smaller. The impact of F_2 is hardly noticeable, but then suddenly at the resonant energy, close to 10.37 kilo-electronvolts, F_2 , which has its direction 90 degrees to F_1 , increases and then drops off only slowly with increasing photon energy in the shown ranges. In contrast, F prime decreases fairly rapidly and F_1 recovers to being close to F_0 . Note also that the phase of the black arrow, very significantly near the absorption energy, this is simply the angle subtended by the blue and black arrows. This means that here, the scattered radiation becomes out of phase with the incident radiation by this phase angle, resulting in partial destructive interference.

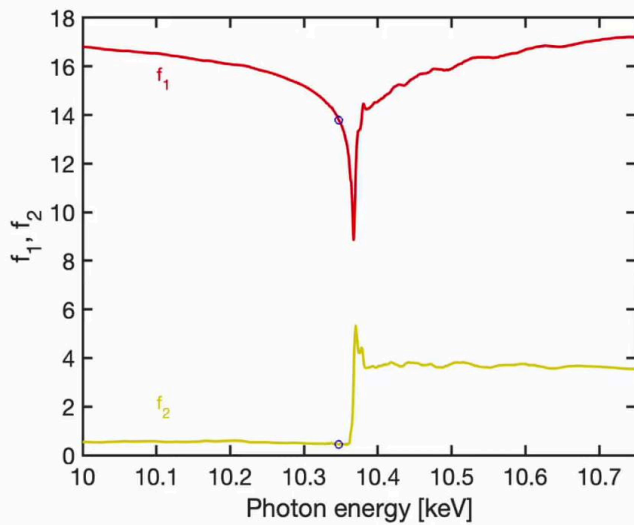
Notes

Summary

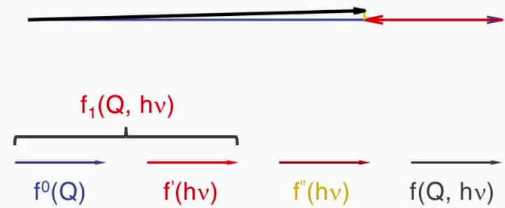
12m 41s



Change in f for Ga @ K-edge and $Q = 4.45 \text{ \AA}^{-1}$



$E = 10.344 \text{ keV}$



In other words, the atom partly absorbs the radiation and the beam is attenuated. Now, do not make the common mistake of thinking that this angle between the total vector sum in black and F_0 in blue means that they propagate in different directions. Differences in angles in the complex plane represent phase differences, not orientation or differences. Before we move on to a quantitative description of the phenomena of reflection, refraction, and absorption of x-rays based on our more complete definition of the atomic form factor discussed in this video, we take a brief look at Compton scattering of high energy photons in the next video.

Notes

Summary

14m 13s

