Quantized form factor shift in the presence of free electron laser radiation

F. Fratini¹,²,³(a), L. Safari², A. G. Hayrapetyan⁴,⁵, K. Jänkälä², P. Amaro⁴,⁶ and J. P. Santos⁶

¹ Universidade Federal de Minas Gerais, Instituto de Ciências Exatas, Departamento de Física
31270-901 Belo Horizonte, MG, Brasil
² Department of Physics, University of Oulu - Fin-90014 Oulu, Finland
³ Institut Néel-CNRS - BP 166, 25 rue des Martyrs, F-38042 Grenoble Cedex 9, France
⁴ Physikalisches Institut, Ruprecht-Karls-Universität Heidelberg - D-69120 Heidelberg, Germany
⁵ Max-Planck-Institut für Physik komplexer Systeme - D-01187 Dresden, Germany
⁶ Centro de Física Atômica, Departamento de Física, Faculdade de Ciências e Tecnologia, FCT,
Universidade Nova de Lisboa - P-2829-516 Caparica, Portugal

received 16 March 2014; accepted in final form 13 June 2014
published online 7 July 2014

PACS 34.80.Qb – Laser-modified scattering
PACS 34.80.Bm – Elastic scattering
PACS 87.64.Bx – Electron, neutron and x-ray diffraction and scattering

Abstract – In electron scattering, the target form factors contribute significantly to the diffraction pattern and carry information on the target electromagnetic charge distribution. Here we show that the presence of electromagnetic radiation, as intense as currently available in free electron lasers, shifts the dependence of the target form factors by a quantity that depends on the number of photons absorbed or emitted by the electron as well as on the parameters of the electromagnetic radiation. As example, we show the impact of intense ultraviolet and soft X-ray radiation on elastic electron scattering by the Ne-like argon ion and by the xenon atom. We find that the shift brought by the radiation to the form factor is of the order of some percent. Our results may open up a new avenue to explore matter with the assistance of laser.

Copyright © EPLA, 2014

Introduction. – Electron scattering is a tool of great importance for exploring the structure of matter [1–6]. The most significant example to highlight such importance is perhaps electron microscopy, where electron scattering is the core process [7]. Electron scattering can be elastic or inelastic. In both cases, the diffraction pattern of the scattered electrons is influenced by the target form factor (FF), also called scattering factor [8,9]. Both elastic and inelastic FFs carry information about the electromagnetic charge distribution of the target [8,9]. FFs are not only relevant in electron scattering, but play an important role also in light diffraction [10] and light absorption [11]. As a consequence, FFs are the subject of research in many scientific areas, such as atomic physics [12], nuclear and subnuclear physics [13,14], crystallography [15] and biology [16,17]. Due to the wide applicability of FFs, exploring the effects of electromagnetic radiation (ER) on the target FFs is of general interest in science.

Laser-assisted electron scattering (LAES) has been the subject of intense research since the 1970s, as a consequence of the development of lasers [18–21]. In this letter we show that the presence of intense ER in electron scattering can be used to control the FFs. More specifically, we show that the presence of ER shifts the argument of the inelastic and elastic FFs from $Q$ to $Q + shk$, where $Q$ is the momentum transfer between electron and target, $k$ is the wave vector of the ER, $h$ is the reduced Planck constant, and $s$ is an integer number that represents the number of photons absorbed (if $s > 0$) or emitted (if $s < 0$) by the electron during the scattering process. A similar linear momentum shift on the whole differential cross-section (DCS) has been already highlighted in the literature [22–25]. However, to the best of our knowledge, the linear momentum shift brought by the ER to elastic and inelastic FFs has not been clearly pointed out in the
in (1) are conserved quantities used to label the electron state inside the ER. However, they do not represent the linear momentum and the energy of the electron (which, on the other hand, are not conserved quantities), as long as the ER is on. They merely identify the electron state inside the ER and are called “linear momentum” and “energy” parameters [32]. Nevertheless, if the ER is switched off, the wave function (1) will become a plane wave and, consequently, $P$ and $E$ will then correctly represent the conserved linear momentum and energy of the electron, respectively. In either cases the ER is on or off, the energy-momentum relation $2mE = P^2$ must be satisfied [34]. Finally, we notice that the parameters $\alpha$ and $\beta$ contain the dependence on the angle between the linear momentum parameter ($P$) and the ER wave vector ($k$).

### Scattering differential cross-section.

Let us now consider LAES by some target characterized by an extended charge distribution and let us denote by $\theta$ the scattering angle. The electron-target interaction is assumed to be electrostatic and the space where the scattering happens is assumed to be filled with linearly polarized ER. We suppose that the ER be non-invasive, i.e., that the target is not perturbed by the ER, as usual in LAES [18]. The amplitude for this scattering process can be written in first-order time-dependent perturbation theory as [34]

$$ A = (i\hbar)^{-1} e \int_{-t}^{t'} dt' \int d^3 r \Psi_{P,k}^*(r,t') V(r) \Psi_{P,k}(r,t), $$

(2)

where $\Delta t = 2t$ is the scattering time interval and the potential $V(r)$ is of the form [8,9,35]

$$ V(r) = \int d^3 \xi \phi_i^*(\xi) V_{ij}(|r - r_\xi|) \phi_j(\xi). $$

(3)

Here $\phi_{i,j}$ are the initial and final target wave functions, while $V_{ij}(|r - r_\xi|)$ denotes the electrostatic potential between two point-like charges at distance $|r - r_\xi|$. We have here assumed, for simplicity, that the target can be described with a single variable $r_\xi$: The extension to many-body targets is trivial. The amplitude (2) does not fully take into account the bound structure of the target. Rather, the target is treated as a source of potential. The second-order scattering term should be included in order to fully account for the target bound structure and resonances. Consequently, the validity range of (2) is restricted to electron energies far from target resonances.

The scattering amplitude (2) may be further manipulated by employing the Jacobi-Anger expansion of the exponentials. The evaluation of the amplitude $A$ proceeds straightforwardly and is similar to the standard evaluation in the absence of ER, as showed, for example, in refs. [8,9]. Mathematical details are given in the supplementary material [36]. On squaring the amplitude, extending the scattering time interval to infinity for obtaining energy conservation, multiplying by the density of final states and normalizing to the electron flux, we obtain...
the differential cross-section for electron scattering in the presence of linearly polarized ER as
\[
\frac{d\sigma^2}{d\Omega}(P_i, P_f) = \left| \frac{P_f}{P_i} \right| J_0^2(\alpha_i - \alpha_f) \left( \frac{2m\hbar c\alpha_i}{(Q + \hbar\omega)^2} \right) \times \left| F(Q + \hbar\omega) \right|^2,
\]
where \( J_0 \) is the Bessel function of order \( 0 \), \( \alpha \) is the previously defined quantity \( \alpha \) calculated for \( P \rightarrow P_i(f) \), while \( \alpha_e \) is the electromagnetic coupling constant.

The quantity \( F(Q) \) is the target FF and reads
\[
F(Q) = \int d^3r e^{i\cdot\cdot\cdot Q}\phi^*_i(r)\phi_f(r).
\]

In elastic scattering, we have \( \phi_i = \phi_f \), and, consequently, the FF represents the Fourier transform of the target charge distribution. In this case, the FF is referred to as the elastic FF. On the other hand, if the scattering is inelastic, we have \( \phi_i \neq \phi_f \), and the FF is referred to as the inelastic FF [8]. In deriving eq. (4), we have furthermore assumed \( \beta_i - \beta_f \approx 0 \), which is justified within the non-relativistic assumption. Here \( \beta_i(f) \) is the previously defined quantity \( \beta \) calculated for \( P \rightarrow P_i(f) \).

In the case the target is an isolated atom, the scattering cross-section gets contributions from both the nucleus and the atomic electrons, for which the relative form factors add coherently but with opposite sign. For non-relativistic energies, we may set the nuclear form factor equal to the nuclear charge (\( Z_n \)). Therefore, in this case eq. (4) reads
\[
\frac{d\sigma^2}{d\Omega}(P_i, P_f) = \left| \frac{P_f}{P_i} \right| J_0^2(\alpha_i - \alpha_f) \left( \frac{2m\hbar c\alpha_i}{(Q + \hbar\omega)^2} \right) \times \left| Z_n - F_{\text{At}}(Q + \hbar\omega) \right|^2,
\]
where \( F_{\text{At}}(Q) \) is the atomic form factor. If the ER is switched off (which is accomplished by setting \( \alpha_{i,f}, k \rightarrow 0 \)), eq. (6) turns out to be equal to the Mott formula [8], as expected.

Since eq. (4) has been derived in non-relativistic first-order perturbation theory, it is not directly applicable when the electron energy is relativistic (\( E_{i,f} \gtrsim 100 \text{ keV} \)) or when the scattering angle is particularly small (\( \theta \lesssim 2^\circ \)). However, relativistic, screening and spin corrections can be added in a similar way as done for the scattering cross-section in the absence of ER, so as to make it applicable for a wider energy range and for small scattering angles [7,9].

By analyzing eq. (4), we notice that the energies of the scattered electrons turn out to be spanned by multiples of the photons energy. The integer number \( s \) is thus easily interpreted as the number of photons absorbed (if \( s > 0 \)) or emitted (if \( s < 0 \)) by the electron during the scattering process, as normally done in LAES experiments [19].

Most importantly, we notice that the presence of ER shifts the argument of the FF from \( Q \) to \( Q + \hbar\omega \). The photon number \( s \), on which the shift depends, can be measured by detecting the kinetic energy of the scattered electron. Due to the fact that such shift does not depend on the ER polarization, it will hold for any kind of ER polarization and, also, for unpolarized light.

It must be underlined that eq. (4) is different from eq. (210) of ref. [20], as our \( Q \) corresponds to \( Q_N \) in ref. [20].

**Kroll-Watson formula re-visited.** – The Kroll-Watson formula (KWF) can be obtained either within the Born approximation, which is in line with our derivation, or within the low-frequency limit (\( \hbar\omega \ll eP \cdot A_0/m \)).

In the light of this, we expect to recover the KWF as a special case of our theory, in the case of elastic scattering, within the assumptions of the first Born approximation. Indeed, if the momentum carried by the absorbed or emitted photons is much lower than the momentum transfer between electron and target, we may use \( Q + \hbar\omega \approx Q \).

Furthermore, the non-relativistic assumption allows for the replacement \( P_{i,f} \cdot k - m\omega \approx -m\omega \) in the denominators of \( \alpha_{i,f} \). Employing these two approximations in (4) yields the KWF for the differential cross-section
\[
\frac{d\sigma^2}{d\Omega}(P_i, P_f) \simeq \left| \frac{P_f}{P_i} \right| J_0^2(\eta) \frac{d\sigma^2}{d\Omega}(Q),
\]
where \( \eta = -eQ \cdot A_0/(\hbar\omega m) \). Kroll and Watson obtained eq. (7) by analyzing the Green function that governs the LAES process. Our result (4) can be thus considered a refinement of the KWF which allows to grasp the effect that the ER brings to the target FF.

The KWF has been widely investigated in atomic physics and has been found to adequately describe the data in several experimental cases (see ref. [19] for a brief account). However, it has been shown that the KWF is unable to describe LAES when the scattering angle is small (\( \theta \approx 9^\circ \)) [38] and when the laser polarization is orthogonal to the linear momentum transfer (\( A_0 \perp Q \)) [39]. Although a few attempts to solve the discrepancies have been made [40–43], the problem is still open [44]. On account of this, we ought to say that the validity of (4) may share the same limitations of KWF.

**Atomic examples.** – Equation (4) is the main result of this letter and describes the differential cross-section for LAES. Specifically, we read from eq. (4) that, in the presence of ER, the target FFs and the differential cross-section depend not only on the momentum transfer \( Q \), but also on the ER wavelength and direction, as well as on the number of photons absorbed or emitted during the scattering process. Thus, these three last parameters can be suitably tuned, in experiments, so as to alter the shape of the target FF to be measured, for a given \( Q \). In the following, we discuss two examples to better highlight the impact of ER on the DCS and on the target FFs.
Fig. 1: (Color online) Differential cross-section for elastic electron scattering by Ne-like argon in the presence of intense ultraviolet ER with wavelength $\simeq 224$ nm. The electron initial energy is $E_i = 2.24$ eV. The ER has direction orthogonal to the scattering axis, linear polarization parallel to the linear momentum transfer, and intensity $I \simeq 10^{15}$ W/cm$^2$. The green short-dashed line refers to electron scattering in the absence of ER, while the crosses represent the best values from experimental measurements taken in absence of ER [45]. The curves denoted by KW are obtained with KWF. $s$ represents the number of photons absorbed (if $s > 0$) or emitted (if $s < 0$) by the scattered electron.

Let us consider elastic electron scattering by an isolated Ne-like argon ion, in the presence of intense ultraviolet ER of wavelength $\simeq 224$ nm (the wavelength of HeAg lasers). In this energy range, the ER may not excite the target, since its first excitation line is at $\sim 4.95$ nm. The target is thus unaffected by the ER, as hypothesized. In fig. 1, we show the differential cross-section, $d\sigma_s/d\Omega$, for such a process, where electron energy and ER parameters are specified. The experimental data agree well with the theoretical predictions for angles $\theta \lesssim 120^\circ$. The theoretical predictions are given by eq. (6) with $\alpha_i, f, k \to 0$, which equals the Mott formula. This also demonstrates that the free Coulomb wave function assumed in eq. (1) is suitable for describing the scattering of electrons by Ne-like argon as provided in the example. For larger angles, there are discrepancies caused by the fact that the slow scattering electron probes the bound structure of the ion, which is not taken into account by the Mott formula, as underlined in introducing eq. (2). Nevertheless, such discrepancies can be removed if Hartree-Fock calculations are used [45].

In fig. 2, we show the atomic FF of a Ne-like argon target in the presence of a soft X-ray ER of wavelength $\simeq 6.2$ nm. The parameter $\tau$ represents the angle between the electron-target linear momentum transfer and the ER direction, i.e. $\cos \tau = k \cdot Q/(|k||Q|)$. For the parameter $s$, see fig. 1.

In fig. 3, we show the atomic FF of a Xe atom in the presence of ER with wavelength $\simeq 6.2$ nm. See figs. 2 and 1 for the parameters $\tau$ and $s$.

Greater with soft X-ray ER, radiation intensities as high as $I \gtrsim 10^{16}$ W/cm$^2$ must be employed, which might generate two-photon absorption peaks. Even so, since the first ionization threshold of Ne-like Ar is at $\sim 2.94$ nm, two-photon absorption may not cause ionization of the target. Such intensities in the soft-X ray regime are nowadays achievable by using XFEL sources [26–28].

The investigation of the linear momentum shift in the FF of neutral samples can also be done, although same care is needed. In order to obtain a considerable shift in neutral atoms, one needs high-intensity radiation whose photons have energy higher than few hundreds electronvolts. Under these conditions neutral samples will undergo several ionizations, leading to numerous background events. However, control parameters can be used to monitor the sample damage, as done in crystallography for...
monitoring the crystal damage [47]. Alternatively, one may use FEL sources and molecular structures. In fact, it has been proved very recently that FEL prevents sample damage in protein structures by simply outrunning it [48]. In view of these considerations, we present in fig. 3 the shift brought to the FF of neutral Xe by soft X-ray ER of wavelength $\sim 6.2$ nm (same as in fig. 2). The FF shift is larger than in fig. 2, although still of the order of some percent. The choice of the Xe atom is motivated by a recent LAES experiment carried out using $fs$ pulses with wavelength of about 800 nm, intensity $10^{12} \text{W/cm}^2$ [21].

The problem of sample damage on a neutral sample might be also circumvented by further increasing the ER intensity so as to reach a new stability regime (stability given by super-intense laser pulses) [49,50].

**Remarks.**—Linear momentum shifts in form factors of clusters and mesoscopic objects can also be studied with the very same formalism developed here [51–54]. A FF shift of the order of $eV/c$ is already significant in such systems, due to their larger size. This can be achieved by using slow electrons and visible light.

As for the case of the KWF [55], the present formalism may be extended to a relativistic framework by employing Volkov solutions and the full electron-target electromagnetic interaction [34]. Such an extension should permit to measure electric and magnetic FFs values at zero argument from scattering events whose momentum transfer is not zero. This would help solve the puzzle arisen from recent measurements on the proton structure, where it has been showed that the ratio between electric and magnetic FFs, at $Q \approx 0$, is slightly less than what is to be expected from QCD considerations [56,57]. The presented formalism might be also employed to shed light onto recent disputed measurements of the proton radius [29,58].

Finally, form factor shifts can be also studied with both radiation-free [59] and radiation-assisted [60] twisted electrons. Such studies will be addressed in future publications.

***

FF acknowledges support by Fundação de Amparo à Pesquisa do estado de Minas Gerais (FAPEMIG) and Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq). LS, KJ and FF acknowledge support by the Research Council for Natural Sciences and Engineering of the Academy of Finland. AGH acknowledges the support from the GSI Helmholtzzentrum and the University of Heidelberg. PA acknowledges support by the German Research Foundation (DFG) within the Emmy Noether program under Contract No. TA 740 1-1. JPS and PA acknowledge support by FCT —Fundação para a Ciência e a Tecnologia (Portugal), through the Projects No. PEstOE/FIS/UI0303/2011 and PTDC/FIS/117606/2010, financed by the European Community Fund FEDER through the COMPETE —Competitiveness Factors Operational Programme. FF is thankful to Prof. MARCELO FRANÇA SANTOS, Prof. SERGIO SCOPETTA, Dr. WEI CAO and Dr. ABRAHAM KANO for useful discussions and comments.

**REFERENCES**

[1] Courttoy A., Fratini F., Scopetta S. and Vento V., Phys. Rev. D, 78 (2008) 034002.
[2] Kamal K. Seth, Dobbs S., Metreveli Z., Tomaradze A., Xiao T. and Bonvicini G., Phys. Rev. Lett., 110 (2013) 022002.
[3] Katsnelson M. I. and Geim A. K., Philos. Trans. R. Soc. A, 366 (2008) 195.
[4] Gargioni E. and Grosswendt B., Rev. Mod. Phys., 80 (2008) 451.
[5] Kamara A. and Ghosh B., J. Appl. Phys., 109 (2011) 024501.
[6] Werner W. S. M., Novák M., Saliván-Pujol F., Zemek J. and Jiříček P., Phys. Rev. Lett., 110 (2013) 086110.
[7] Williams D. B. and Carter C. B., Transmission Electron Microscopy: A Textbook for Material Science (Springer) 2009, Chapt. 2 and 3.
[8] Fultz B. and Howe J. M., Transmission Electron Microscopy and Diffractionmetry of Materials (Springer) 2008, Chapt. 3 and 4.
[9] Povh B., Rith K., Scholz C. and Zetsche F., Particles and Nuclei: An Introduction to Physical Concepts (Springer) 2006, Chapt. 5.
[10] Chantler C. T., J. Phys. Chem. Ref. Data, 29 (2000) 597.
[11] Cesareo R., Hanson A. L., Gigante G. E., Pedriza L. J. and Maitaboally S. Q. G., Phys. Rep., 213 (1992) 117.
[12] Alatas A., Said A. H., Sinn H., Bortel G., Hu M. Y., Zhao J., Burns C. A., Burkel E. and Alp E. E., Phys. Rev. B, 77 (2008) 064301.
[13] Frosch R. F., McCarthy J. S., Rand R. E. and Yearian M. R., Phys. Rev., 160 (1967) 874.
[14] Bernauer J. C. et al., Phys. Rev. Lett., 105 (2010) 242001.
[15] Maslen E. N., Fox A. G. and O’Keeffe M. A., in International Tables for Crystallography, Vol. C: Mathematical, Physical and Chemical Tables (Kluwer Academic Publishers, Dordrecht) 1992, sect. 6.1.1; Colliex C. et al., in International Tables for Crystallography, Vol. C: Mathematical, Physical and Chemical Tables (Kluwer Academic Publishers, Dordrecht) 1992, sect. 4.3.1.
[16] Tartari A., Taibi A., Bonifazzi C. and Baraldi C., Phys. Med. Biol., 47 (2002) 163.
[17] Morin L. R. M., J. Phys. Chem. Ref. Data, 11 (1982) 1091.
[18] Mason N. J., Rep. Prog. Phys., 56 (1993) 1275.
[19] deHarak B. A., Ladino L., MacAdam K. B. and Martin N. L. S., Phys. Rev. A, 83 (2011) 022706.
[20] Ehlotzky F., Jarůn A. and Kamiński J. Z., Phys. Rep., 297 (1998) 63.
[21] Kanya R., Morimoto Y. and Yamanouchi K., Phys. Rev. Lett., 105 (2010) 123202.
