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Phys. Rev. Lett. **108**, 063007 — Published 10 February 2012

DOI: 10.1103/PhysRevLett.108.063007

Angle-resolved electron spectroscopy of laser-assisted Auger decay induced by a few-femtosecond x-ray pulse

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Two-color (x-ray + infrared) electron spectroscopy is used for investigating laser-assisted KLL Auger decay following $1s$ photoionization of atomic Ne with few-femtosecond x-ray pulses from the Linac Coherent Light Source (LCLS). In an angle-resolved experiment, the overall width of the laser-modified Auger electron spectrum and its structure change significantly as a function of the emission angle. The spectra are characterized by a strong intensity variation of the sidebands revealing a gross structure. This variation is caused, as predicted by theory, by the interference of electrons emitted at different times within the duration of one optical cycle of the infrared dressing laser, which almost coincides with the lifetime of the Ne $1s$ vacancy.

Auger decay is the dominating relaxation mechanism for ionized atoms of low nuclear charge with inner shell vacancies and is a salient and one of the most fundamental processes arising from the interaction of short wavelength radiation with matter. Since this process takes place on the ultra-short timescale of a few femtoseconds, information about its dynamics is obtained, in general, indirectly via spectroscopic studies and comparison with advanced theoretical models (e.g. [1]). Alternatively, direct measurements in the time domain have become possible by using ultra-short attosecond pulses produced by harmonic generation from a few-cycle femtosecond laser [2–4]. Recently, new opportunities were opened up by the advent of free-electron lasers (FELs) operating in the XUV and x-ray wavelength regime [5–7]. The unprecedented properties of these new sources, providing very intense femtosecond XUV and x-ray pulses, have given rise to new results arising from the investigation of the atomic inner-shell phenomena, especially the atomic Auger process [8, 9].

In particular, various two-color experimental schemes have been explored to study atomic photoionization [10], as well as, atomic and molecular relaxation processes [11]. For the case of overlapping infrared (IR) and x-ray pulses, the strong optical field gives rise to sidebands in the electron spectrum. These sidebands arise due to interaction of the photo- or Auger electrons with the laser field and are associated with absorption or emission of a few IR photons by the outgoing electrons. For the decay of an inner shell vacancy, this phenomenon is known as laser-assisted Auger decay (LAAD) and was first observed by Schins et al. [12, 13] for LMM Auger transitions in argon. In those experiments the Auger electrons were detected in an angle-integrated way, and the measured spectra consisted of strong central (unperturbed) lines and a few sidebands separated by the IR photon energy. The intensity of the sidebands decreased regularly with the number of excess photons.

In this Letter, we present the first experimental investigation of the angular and spectral distribution of sidebands in LAAD induced by femtosecond x-ray pulses from the Linear Coherent Light Source (LCLS) at SLAC [7]. Our experimental results demonstrate the strong modification of the Auger electron angular distribution by the infrared dressing field, in particular a pronounced modulation of the sideband intensities, which are interpreted as arising from the interference of electron waves emitted within one optical cycle. Thus, the experiment reveals a fundamental process, one which will be observable in all two-color angle-resolved electron studies for temporally overlapping pulses, where the relevant time scale for electron emission (pulse duration for photoionization and core hole lifetime for Auger emission) is on the order of an optical cycle. The phenomenon is independent of the target under investigation, i.e. will be observed for atoms, molecules, clusters or solids. The target chosen for the demonstration is atomic Ne, since the lifetime of the Ne 1s vacancy (about 2.4 fs) is comparable to the duration of one optical cycle of an 800 nm optical laser (about 2.6 fs). In addition, Ne has a rather simple KLL Auger spectrum dominated by the strong transi-

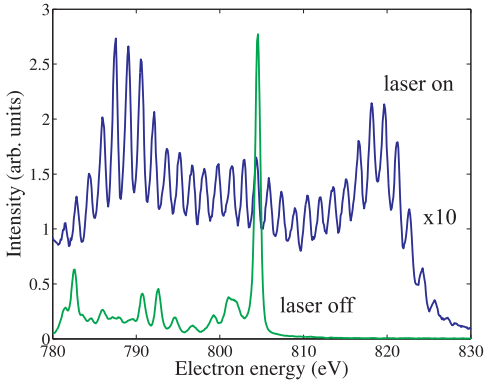

FIG. 1: KLL Auger spectrum of atomic Ne recorded at the emission angle of 0° relative to the polarization vector of the optical field with and without additional optical dressing field.

tion $Ne^+\ 1s^{-1}2p^6 \rightarrow Ne^{2+}\ 2p^4\ ^1D_2$ [14] with the Auger electron energy of 804.3 eV. In the absence of the optical laser the angular distribution of the KLL Auger electrons is isotropic [15] since the 1s vacancy produced by linearly polarized x-rays cannot be polarized (aligned). Thus any anisotropy in the angular distribution of the KLL Auger electrons in LAAD is determined by interaction of electrons with the optical laser field.

The experiments were performed at the AMO (atomic, molecular, and optical physics) endstation at the LCLS [16]. Neon gas was ionized by x-ray pulses from LCLS operating at a wavelength of 1.2 nm, i.e. 1000 eV photon energy, in the low-charge mode providing extremely short pulses of a few (2-5) femtoseconds duration. The pulses of a Ti:Sapphire optical laser operating at a wavelength of 800 nm were electronically synchronized to the LCLS x-ray pulses [17] and provided a dressing field of about 10^{12} W cm^{-2} in the interaction region. The spatial overlap of the x-ray and IR beams was achieved by viewing the beams on a YAG screen which was moved into the interaction region. Rough temporal overlap was achieved with the help of a fast photodiode providing a time resolution in the order of 30 ps. Precise temporal adjustment and optimization of the spatial overlap was obtained on the two-color signal in the photoelectron spectra. In order to minimize the measurements sensitivity to spatial and temporal fluctuations of the LCLS pulses, the optical laser was operated with long pulses (about 3 ps) compared to the temporal jitter between the x-ray and IR pulses of about 140 fs [17, 18] and was focused to a diameter of about 70 μm in the interaction volume (compared to the 20 μm x-ray focal spot size.) Typical spectra recorded with and without the optical laser are presented in Fig. 1.

Application of the optical dressing field dramatically alters the electron spectrum. In this case, the electron emission is completely governed by the optical field leading to a sideband structure, which extends over more than 50 eV, i.e. containing more than 15 sidebands on each side of the undressed Auger line. This dramatic effect is mainly caused by the high kinetic energy of the Auger electrons, which favors the production of sidebands even at relative small IR intensities. The small contributions from the satellite structure, which is observed on the low kinetic energy side of the main $^1D_2$ line, produces a slight asymmetry in the LAAD spectrum.

The angular distributions of the Auger electrons are analyzed using three time-of-flight (TOF) analyzers, which were mounted at different angles (0°, 54.7° and 90°) with respect to the linear polarization vector of the x-ray pulses. Since for the measurement only the emission angle with respect to the optical laser polarization is important, all other angles were obtained by rotating the relative orientation of the linear polarization vector of the optical laser by means of a half-wave plate. The TOF analyzers have an energy resolution of about 650 meV for 800 eV electrons, i.e. sufficient to resolve the individual sidebands (cf. Fig. 1), which are separated by the IR photon energy (1.55 eV). Typical acquisition times for an electron spectrum were in the order of 20-30 min, i.e. an average of more than 30 000 pulses.

Recent theoretical papers [19–21] have shown that for Auger electrons with large energy, the number of sidebands in the electron spectra is large and the intensity of the sidebands, when measured in an angle-resolved experiment, is modulated revealing a so-called 'gross structure'. Moreover, it was predicted that the number and the intensity of the sidebands strongly depend on the emission angle relative to the optical laser polarization. An angle integrated spectrum masks these effects but gives a result consistent with the earlier observations of [12, 13]. Since the energy of the Auger electron is large, one can use the strong field approximation (SFA) [22] to theoretically describe the Auger-electron spectrum in the IR laser field. This approximation assumes that in the inner atomic region the Auger process is not influenced by the IR laser field, a condition that is satisfied for the moderately strong IR fields considered here. In contrast, in the outer region, the interaction of the emitted electron with the residual ion can be ignored and the propagation of the emitted electron is determined by the IR field only. Thus the outgoing electron can be described by a Volkov wave function [23]. SFA was already successfully applied to the Auger electron emission in Refs [12, 13, 24]. A simplified description of the Auger-electron spectra, based on SFA, has been developed in Refs. [19, 20]. They have demonstrated that for sufficiently large Auger energies the SFA gives results close to the numerical solution of the non-stationary Schrödinger equation which includes both interactions of the electron: the IR field and the residual ion. The theoretical results presented here have been obtained using this model [19, 20].

The strong variation of the LAAD spectra as function of the emission angle is displayed in Fig. 2. The electron spectrum recorded with the detector set perpendicular to the polarization of the IR dressing field (90°) shows an unperturbed Ne KLL Auger line. As the angle is reduced, the Auger line splits up into sidebands and for emission parallel to the optical field (0°) the spectrum is dominated by the sideband structure. At high emission angles (50° - 80°) the line intensities are

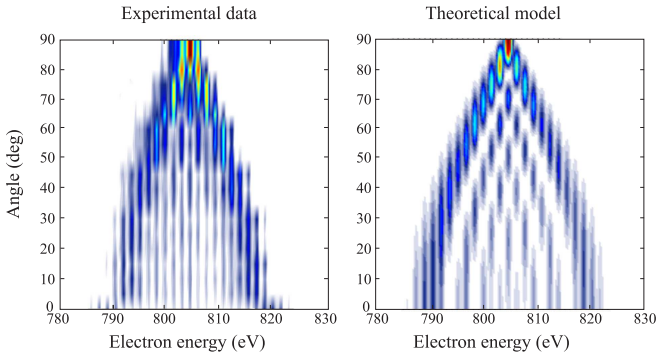

FIG. 2: 2D angle-resolved spectra of KLL Auger electrons from Ne in the laser field presented as a color-scaled plot. Left panel: the observed spectra; right panel: the calculated spectra, obtained for a laser intensity 0.7×10^{12} W cm^{-2}.

clearly characterized by a modulated distribution with strong minima and maxima for particular sidebands. This behavior is revealed even more clearly in the theoretical spectra (Fig. 2, right panel) which were calculated for an optical laser intensity of 0.7×10^{12} W cm^{-2}. The intensity of the central (undisturbed) peak is almost zero for some angles showing that its strength is completely redistributed among the sidebands. In general, both the theory and the experiment show that the most populated groups of sidebands lie on the periphery of the spectra. Theory [21] shows that the energy interval between these most populated groups is $\sim \sqrt{IE_a}\cos\vartheta$, where I is the IR intensity, E_a is the Auger electron energy, and ϑ is the emission angle. The width of the spectral distribution decreases with increasing emission angle. At exactly 90° the theory predicts only one peak since the IR electric field is zero along this direction.

The origin of these intensity oscillations lies in interferences caused by the emission of the Auger electrons at different times during one optical cycle. As shown in Ref. [21] there are two types of interference. For an Auger lifetime that is larger than the period of the IR field, Auger electrons emitted at different periods but at the same phase of the IR field interfere and result in the formation of the sidebands. However another interference arises when emissions occurs at two different times that have the same vector potential within one IR cycle. These electrons acquire the same energy from the IR laser field and the corresponding electron waves can therefore interfere at the detector position. It is this interference that leads to a modulation of the sideband amplitudes (see [21] for details).

This gross structure is evident in both the theoretical and experimental spectra for large emission angles. Figure 3 shows the measured and calculated spectra for an emission angle of 60°. In order to obtain good agreement with the experiment the theoretical spectrum includes averaging over a small intensity interval (0.5-0.8)$\times 10^{12}$ W cm^{-2}. This averaging takes into account that the spatial intensity distribution in the focus is not flat but varies with position. The average calculated spectrum is in excellent agreement with the experiment. In the central part of the spectrum, the relative intensity of neighboring sidebands oscillates between minimal and maximal values, proof positive that interference between electron waves emitted at different times during a single optical cycle is occurring as predicted. In addition, the observation of these interferences evidences that the Auger electrons are emitted coherently independent from the exact moment of emission during the decay, which is consistent with the theoretical treatment [25]. We note, that due to the temporal jitter between the FEL and IR pulses, the experimental spectra were recorded at random time delays and phase differences. This jitter was theoretically simulated by averaging over the time-delay between IR and x-ray pulses within one optical cycle. All calculations were performed for a single Auger transition with no contribution from weaker satellite lines [26]. These satellites are responsible for the higher intensity in the low energy part of the experimental spectrum. However, the slight asymmetry between low and high-energy parts of the calculated spectrum is inherent in the theory (see eqs. (20) - (22) in [25]).

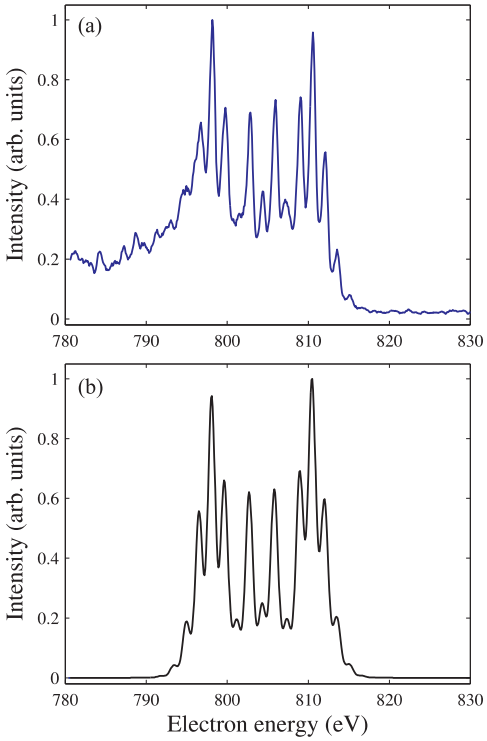

FIG. 3: Experimental (a) and theoretical (b) Auger spectra for the emission angle of 60 degree.

The result of the non-uniform IR intensity is more severe for emission angles close to 0° (Fig. 2), as revealed by the differences between the experimental and the "ideal" theoretical spectrum. At small angles the central region of the experimental spectrum is nearly uniform, i.e. the intensity varies only slightly from line to line. In contrast, the theoretical spectrum, calculated for a constant IR intensity, shows a rather strong modulation. In figure 4 we show the experi-

mental and calculated spectra for electrons emitted at $\vartheta = 0°$. In the measured spectrum (Fig 4a), the increased intensity for the two groups of high-order sidebands near 820 and 790 eV is clearly observed, but in other regions the variation is small. However, the theoretical spectrum, solid curve in Fig. 4c, calculated for an IR field intensity of 1.0×10^{12} W cm^{-2} shows a well developed modulation also in the central part of the distribution. This structure is less apparent in the experimental spectrum. To check our assumption of a non-uniform IR field causing this discrepancy we performed simulations for different intensities in the range (0.6 - 1.0)$\times10^{12}$ W cm^{-2}. As an example in Fig. 4c, two spectra for the intensities 1.0×10^{12} W cm^{-2} and 0.7×10^{12} W cm^{-2} are shown. One clearly sees that in many cases a particular sideband has minimal or maximal intensity changing from one spectrum to the other. The strong dependence of the gross structure on the IR laser intensity is related to the fact that the former is determined by the above mentioned parameter $\sqrt{IE_a}$. At large kinetic energies of the Auger electron (E_a) even a small variation of the intensity (I) leads to a noticeable effect in the sideband structure. The spectrum averaged over the IR intensity interval (0.6-1.0)$\times10^{12}$ W cm^{-2} is displayed in Fig. 4b. For the average calculated spectrum the gross structure in the middle part almost disappears and the spectrum becomes very similar to the experiment. The strong intensity dependence of the positions of maxima and minima of the gross structure, especially in the central part of the spectrum, leads to a smoothing of the interference patterns.

In conclusion, we have observed pronounced interference pattern (gross structure) in the sideband structure produced by Auger electron emission in a laser field. The Auger spectra depend strongly on the emission angle and the angular dependence is entirely determined by the interaction of the outgoing electron with the optical dressing field. The observation and investigation of this new phenomenon was enabled by the unique characteristics of novel free electron laser sources, such as LCLS, delivering high photon energy and short pulse durations and providing the possibility to perform high-resolution electron spectroscopy. The experimental results are in good agreement with the theoretical predictions based on SFA. This study provides a foundation for probing the dynamics of Auger electrons in the presence of a strong IR field and eventually in the future of the Auger process itself, in particular by using different wavelengths of the dressing field chosen to match at best the Auger lifetimes under investigation. In addition, the interference pattern is quite sensitive to the IR laser intensity and this feature might therefore be used for characterizing the field structure at the focus of the IR laser pulse. Finally, the existence of the gross structure and its sensitivity to the IR field intensity show that great care has to be taken in the interpretation of electron angular distribution data in high resolution pump-probe experiments.

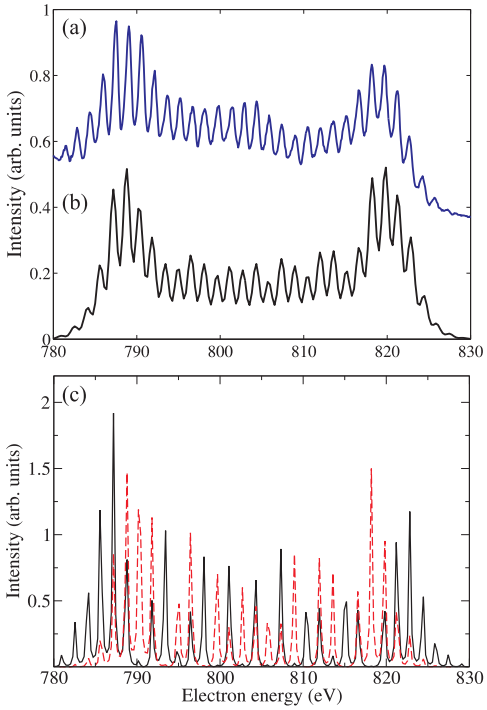

FIG. 4: (a) Experimental Auger spectrum recorded at 0° relative to the IR linear polarization direction. (b) Calculated Auger spectrum averaged over the intensity interval (0.6 - 1.0)$\times10^{12}$ W cm^{-2}. (c) Calculated Auger spectra for two IR intensities: 1.0×10^{12} W cm^{-2} (solid curve) and 0.7×10^{12} W cm^{-2} (dashed curve).

Portions of this research were carried out at the Linac Coherent Light Source (LCLS) at the SLAC National Accelerator Laboratory. LCLS is funded by the U.S. Department of Energy Office of Basic Energy Sciences. It is our pleasure to acknowledge the work of the scientific and technical teams at LCLS. N.M.K. is grateful to the Donostia International Physics Center (DIPC) for hospitality extended to him during his visit. K.U. acknowledges the support by MEXT. A.R.M. acknowledges financial support from the Munich-Centre for Advanced Photonics (MAP) and is particularly grateful to F. Grüner. J.C. acknowledges support from SFI Grant No. 07/IN.1/I1771. G.D. and C.R. acknowledge support from DOE/BES under contracts DE-AC02-06CH11357 and DE-FG02-04ER15614. LFD acknowledges support from NSF PHY-1004778.

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