Observation of Laser-Assisted Auger Decay in Argon

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The effect of an intense laser field on the energies of Auger electrons has been investigated using electron spectroscopy. A laser-generated gallium plasma pumped by a subpicosecond laser yields pulsed broadband x radiation, used to induce *LMM* Auger transitions in argon. The intense dressing beam ($(\lambda = 800$ nm) causes the appearance of up to four sidebands on both sides of the original Auger lines. Recording the magnitude of the sidebands versus the time delay between the dressing and x-ray pulses shows that our source delivers subpicosecond pulses in the 250–400 eV range.

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Optical transitions between continuum states can easily be driven into saturation. This was first seen in multiphoton ionization (MPI), where the strong coupling in the continuum resulted in the appearance of a multitude of peaks in the photoelectron spectrum, spaced by the photon energy. This effect, known as above-threshold ionization (ATI) [1], has attracted considerable theoretical attention, because it offers an example of nonperturbative behavior [2].

In this Letter we present the observation of a new process, laser-assisted Auger decay (LAAD), which is another manifestation of strong continuum coupling: An Auger decay that takes place in an optical radiation field can be accompanied by absorption or emission of a number of photons. This results in the appearance of "sidebands" on the Auger peak in the electron spectrum.

Nonperturbative behavior, where several sidebands of different order become similar in magnitude, occurs for this process at light intensities much lower than those required for nonperturbative ATI, as will be explained below. In addition, laser-modified Auger decay has the advantage that the initial ionization event has a very simple theoretical description compared to the high-order multiphoton step in ATI. This makes LAAD a good candidate for study of continuum transitions.

Auger decay [3] is a well-characterized effect resulting from inner-shell vacancies. For instance, if an inner-shell electron is removed by photoionization, the core-hole ion may decay through a process in which one valence electron fills the vacancy while a second one is ejected into the continuum with a well-defined energy, independent of the incoming photon energy [4]. This effect has been extensively studied during the past few years, especially in rare gases, using synchrotron sources, and the Auger lines are well documented [5].

In a full quantum-mechanical treatment the absorption (emission) of photons in a final continuum state is described as stimulated (inverse) bremsstrahlung (see [6] and [7]). Although such a description of laser-assisted x-ray photoionization (a process very similar to LAAD) is available [8], an estimate of the light intensity needed to get observable LAAD can be more readily obtained from the purely classical model known as the simpleman's theory [9]: An electron with an initial kinetic energy U_a corresponding to the Auger decay is introduced in the optical field $E_0\cos(\omega t)$ at a time t_0 and from then on moves as a free point charge. The electron average kinetic energy in the field is then easily shown to be

$$U \approx U_a - \sqrt{8U_aU_p}\sin(\omega t_0)\cos\theta + U_p[1 + 2\sin^2(\omega t_0)], \tag{1}$$

where U_p is the quiver energy of the electron in the field (ponderomotive energy [10]) and θ is the angle between the light polarization and the velocity of the Auger electron. Since in our case $U_p \ll U_a$, we can ignore the last term in Eq. (1). This simple approach predicts that the initial energy U_a can be increased or decreased (depending on t_0) by an amount $\Delta U = \sqrt{8U_aU_p}$. In reality this energy change will be quantized as a multiple of the photon energy, resulting in a sideband structure rather than a continuous broadening of the Auger line. The classical argument will still yield the correct prediction for the energy range spanned by these sidebands, leading to

$$N = 2\Delta U/\hbar\omega = 2\sqrt{8U_aU_p}/\hbar\omega \tag{2}$$

sidebands around the zero-field line. As a consequence, N scales as the square root of the intensity. Furthermore,

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since U_a is typically much larger than U_p, multiple sidebands occur at very modest values of U_p as compared to ATI.

Experimental study of LAAD is also attractive for other reasons. A measurement of the lowest-order LAAD sideband in the perturbative regime as a function of the timing of the optical pulse provides a measurement of the temporal profile of the x-ray pulse. The Auger decay occurs instantaneously on the time scale of the laser pulse (as can be inferred from the Auger linewidths [5]), and this makes the sideband signal proportional to the cross correlation between the optical and the x-ray pulses. Furthermore, an experimental advantage is that the Auger energy is independent of the x-photon energy, so that broadband x radiation may be used.

This Letter reports the first demonstration of LAAD. The $L_{23}M_{23}M_{23}$ transition in argon is dressed by 1.55 eV infrared (IR) photons from a femtosecond intense titanium:sapphire (Ti:S) laser. Photons with energies in the range from 250 eV (argon L edge) to 400 eV, where the argon photoabsorption cross section decreases exponentially, eject an electron from the $2p^6$ shell. The resulting core-hole ion decays mainly into the ground-state configuration of the doubly charged ion ($2p^6 3s^2 3p^4$) by ejecting a second electron with an energy of about 200 eV. Because of the splitting of the initial ($^2P_{3/2}$, $^2P_{1/2}$) and final (^1S, ^1D, $^3P_{0,1,2}$) states, the Auger line has a complicated structure of partially overlapping peaks, resulting in four resolvable components [11].

Figure 1 shows a schematic of the experimental setup. The output beam of a self-model-locked Ti:S oscillator is stretched to 400 ps and amplified in two stages. After optimum recompression one obtains 40 mJ pulses of 150 fs at 800 nm with a repetition rate of 10 Hz and a near-Gaussian spatial profile of 10 mm FWHM [12]. A beam splitter divides the incoming beam into two branches. One of the beams is used to generate the strong optical field. This "dressing" beam is led through a variable delay and passed through a multilayer dielectric mirror. By slightly rotating this mirror the intensity could be adjusted. Subsequently the beam is focused onto an electron spectrometer by means of a planospherical lens ($f = 2$ m) where it crosses the x-ray beam at a right angle. The pulse energy was monitored by recording the reflection from the lens on a photodiode.

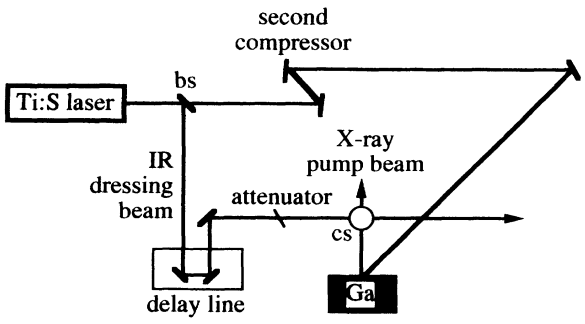

FIG. 1. Schematic of the experimental setup. An amplified Ti:S laser delivers pulses that are divided by a beam splitter "bs." The x-ray pump beam passes through a grating-pair compressor before it is focused on the gallium target. The plasma radiates x-ray pulses which traverse the interaction region marked "cs." The other beam is led through a delay line before being focused into the interaction region of an electron spectrometer.

The time-of-flight electron spectrometer is of the magnetic bottle type and is designed for collection over 2π sr solid angle [13]. An aperture placed inside the flight tube limits the acceptance volume of the spectrometer to a "sensitivity cylinder" of about 2 mm length extending along the spectrometer axis and 0.25 mm diameter. The focusing lens is tilted by 30° to create an astigmatic focus, making it possible to envelop the sensitivity volume of the spectrometer completely with the dressing beam. The overlap is optimized by maximizing the MPI signal obtained from xenon gas. The lens is positioned at such a distance from the spectrometer that, in the direction perpendicular to the spectrometer axis, the beam measures approximately 0.5 mm FWHM. As a consequence, the intensity at the focus never exceeded 10^{12} W/cm^2, and indeed no photoelectrons from argon were detected with the optical pulse alone.

The second beam from the beam splitter is used to produce the x rays by pumping a laser-generated plasma. It is passed through a single-pass grating compressor to allow for independent variation of the pulse duration of the x-ray pump and IR dressing beams. By means of a lens of 200 mm focal length, the pump beam is focused onto a gallium target placed 25 mm below the spectrometer axis, thereby creating a plasma burst which emits ultrashort broadband x radiation. An energy of 3 to 4 mJ is used. It corresponds to a peak intensity on the target of 10^{16} to 10^{17} W/cm^2.

The x rays are sent to the spectrometer sensitivity region via two apertures separated by 10 mm, the second one being a small (2×0.4 mm) slit. This slit was found to improve the signal quality by shadowing the sensitivity region from most of the expanding plasma. The target is slightly heated to keep the gallium liquid. This makes the surface self-regenerating, while the low vapor pressure (below 10^{-9} Torr) is compatible with our ultrahigh vacuum system.

The target gas (argon) is fed into the spectrometer through a pulsed valve, with a backing pressure of about 5 bars. Working conditions imply a mean pressure of 10^{-5} Torr in the interaction volume, peaking to about 10^{-1} Torr for less than a millisecond, synchronized with the laser pulse. The photoelectrons are detected by a multichannel plate located at 1.50 m from the interaction zone and are analyzed according to their time of flight by a digital sampling scope. To obtain sufficient resolution (0.5 eV) around 200 eV, the electrons are retarded by 194 eV immediately after leaving the interaction volume.

In order to study the intensity dependence of the LAAD spectra, the compressor in the pump branch was set for the shortest possible pulse duration (0.5 ps). The dressing pulse was made much longer (2.5 ps) by adjusting the compressor of the Ti:S laser. The LAAD signal was optimized with respect to the delay, which resulted in a situation where all Auger decays occur near the peak of the optical pulse, at essentially the same light intensity. Figure 2(a) shows typical electron energy spectra for different values of the light intensity. The absolute intensities have been calculated from the beam parameters and are accurate to within 50%. The relative intensities follow directly from the monitored photodiode signal, accurate to within 1%. As mentioned, the spectrum without probe ($I = 0$) consists of four peaks of approximately 1 eV width. At higher dressing-field intensities all of these four peaks decrease, while sidebands appear. For instance, at 6×10^{10} W/cm^2, the sideband at 202.3 eV has grown bigger than its neighboring zero-field peak at 201.3 eV. At 1.4×10^{11} W/cm^2 the latter peak has disappeared completely and a second sideband is detected to the left of it.

According to Eq. (1), the sideband structure of a single Auger line depends in principle on the angular distribution of the ejected electrons and on their energy. In our case, the energy differences between the four components are negligible. If we also assume identical angular distributions, the complex structure of the Auger transition itself can be eliminated by deconvoluting the raw spectra with the zero-field spectrum. Note that the unpolarized nature of the x-rays results in some averaging over angles and therefore helps in justifying this assumption.

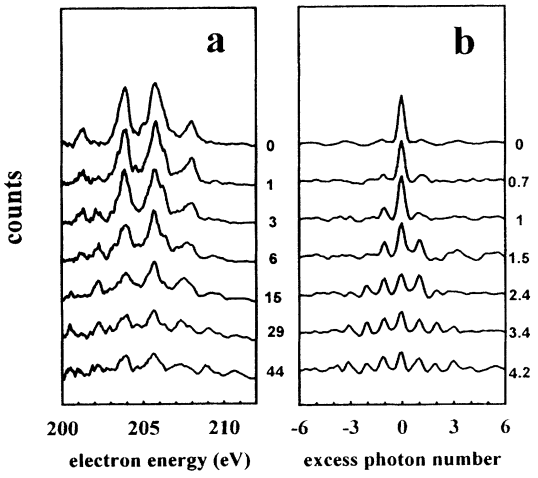

FIG. 2. The traces on the left show the LAAD electron spectra at different dressing field intensities. They are labeled with the experimental intensities of the dressing optical field, in units of 10^{10} W/cm^2. The traces on the right dispaly the corresponding deconvoluted spectra showing the LAAD sidebands of a single Auger line. They are labeled with the number of sidebands expected according to Eq. (2) in the text.

The results of the deconvolution are shown in Fig. 2(b), which thus displays the sideband structure of a single Auger line. The expected number of sidebands can be estimated according to Eq. (2), yielding $N = 6.3 \times 10^{-6}\sqrt{I(\mathrm{W/cm^2})}$. As can be seen from Fig. 2(b), the predictions from the simpleman's theory (labeling the deconvoluted curves) are in excellent agreement with the experimentally observed number of sidebands. A comparison with more elaborated model calculations will be given in a forthcoming paper [14].

A cross correlate between the optical and x-ray pulse has been obtained (Fig. 3) with the shortest possible pulses (150 fs) in both the x-ray pump and IR dressing beams, by scanning the delay between the two pulses. To this end, the compressor in the x-ray pump beam was removed.

To recover the x-ray pulse duration from this data, we assume the probe pulse to be much shorter than the x-ray pulse. The former can then be described by a delta function. The experimental data points are fitted well on assuming the x-ray pulse to be an exponential function of time $e^{-\gamma t}$ and on describing the finite size of the sensitivity area of the spectrometer by a Gaussian function with full width at $1/e$ maximum of 0.24 mm. The $1/e$ decay time of the x-ray pulse is then found to be $\gamma^{-1} = 0.7$ ps. For the present setup with crossed beams, the time resolution of the method is in fact limited by the transit time through the area of sensitivity (0.8 ps). The measured x-ray pulse duration is consistent with streak-camera measurements taken under amplified-spontaneous-emission–free, dye-laser plasma excitation [15] and of earlier x-ray or visible cross-correlation experiments [16]. The resolution of the pump-probe measurement is about 5 times better than for the streak camera and could still be improved to 100 fs by switching to a collinear setup.

In conclusion, we have demonstrated for the first time the occurrence of laser-modified Auger decay, in good agreement with the predictions of a simple classical model. This new physical process has been used as an accurate tool for measuring the duration of x-ray

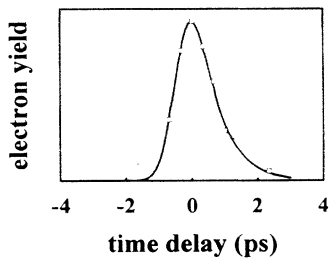

FIG. 3. Cross correlation of the x-ray and optical (infrared) pulses. The datapoints are obtained by integration of the electron counts in a 1 eV region around the sideband centered at 202.3 eV, for different values of the time delay between x-ray pump and IR dressing pulses. The solid line is the fit to the data using an x-ray pulse duration of 0.7 ps.

pulses. Using this technique on x rays emitted by a laser-generated plasma, we measure a subpicosecond pulse for photons emitted in the range of 250 to 400 eV.

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