A quantum mechanical model of the time–resolved Auger measurement

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The theoretical model used for calculating Figs. (3) is a straight forward extension of the theory of the attosecond XUV pulse measurement published in Ref [1]. We generalize Eq. (2) of Ref. [1] in the form

$$b(\mathbf{p}) \propto i \int_{t_0}^{\infty} \exp\left[-i \int_{t'}^{\infty} \frac{1}{2}\left(\mathbf{p}-\mathbf{A}(t'')\right)^2 \mathrm{d}t'' + iW_{\mathrm{kin}}t'\right] \sqrt{\rho(t')}\chi(\mathbf{p}-\mathbf{A}(t'))\mathrm{d}t' \tag{1}$$

Here $b(\mathbf{p})$ is the momentum space wave function after passage of both, the XUV and the laser pulse. W_{kin} is the kinetic energy of the Auger electron without external fields, as defined in the main article. The laser field $\mathbf{E}(t)$ is related to the electron momentum boost $\mathbf{A}(t)$ by $\mathbf{E}(t) = -\partial_t\mathbf{A}(t)$ (in atomic units). The exponential factor in the integrand describes the motion of continuum electrons in the laser field. The difference to the theory presented in [1] is that here the generation of continuum electrons is not by a dipole transition induced by the field $\mathbf{E}$, but by an Auger process, which is described by the factors $\sqrt{\rho(t')}\chi(\mathbf{p}-\mathbf{A}(t')$. The laser field involved is weak and its influence on the Auger process itself can be neglected. Therefore the shape of the electron wave function as it first appears in the continuum is the same as in the field free Auger decay. Using only the dominant configuration of the Auger state and neglecting exchange symmetry, an ansatz for the initial continuum wave function in coordinate space $\chi(\mathbf{r})$ is derived from the known energies and symmetries of the Auger transition by writing

$$\begin{aligned}
&\Phi_{4s}(\mathbf{r}_0)\Phi_{4p}(\mathbf{r}) \rightarrow \Phi_{3d}(\mathbf{r}_0)\chi(\mathbf{r})\\
&\approx \frac{1}{|\mathbf{r}_0-\mathbf{r}|}\Phi_{4s}(\mathbf{r}_0)\Phi_{4p}(\mathbf{r})\\
&\approx \sum_l \frac{4\pi}{2l+1}\frac{r_0^l}{r^{l+1}}P_l(\cos\theta)\Phi_{4s}(\mathbf{r}_0)\Phi_{4p}(\mathbf{r})
\end{aligned} \tag{2}$$

As factor functions $\Phi_{3d}(\mathbf{r}_0)$, $\Phi_{4s}(\mathbf{r}_0)$ and $\Phi_{4p}(\mathbf{r})$ we use the corresponding hydrogen-like functions with an effective charge, which is adjusted to reproduce the known energies. Due to the quadrupole nature of the relaxation process $4s \rightarrow 3d$ only the quadrupole term $l = 2$ of the multipole expansion contributes. It turns out that the integral (1) is rather insensitive to the exact choice of χ, which justifies the approximations employed. The strength of the transition to χ is proportional to the Auger decay rate Γ and to the population ρ of the Auger state. The rate equation for ρ is

$$\frac{d}{dt}\rho(t) = -\Gamma\rho(t) + \alpha I_{\text{xuv}}(t). \tag{3}$$

The Auger state is produced from the neutral atom by the single–photon ejection of the 3d–electrons from the neutral atom at a rate that is proportional to the XUV pulse intensity $I_{\text{xuv}}(t)$. The solution of (3) with initial condition $\rho(t_0) = 0$ is

$$\rho(t) = \alpha \int_{t_0}^{t} I_{\text{xuv}}(t')e^{-\Gamma(t-t')}\mathrm{d}t' \tag{4}$$

The use of a rate equation is justified as long as Auger decay times are much longer than the characteristic time of 3d–electron ejection, i.e. $\Gamma << I_{3d}$. With the ionization potential $I_{3d} \approx 95\,eV$ this inequality is safely fulfilled for decay rates down to $0.2\,fs$. Note that the square root $\sqrt{\rho(t)}$ enters Eq. (1), since the integral is over wave function amplitudes rather than populations.

The three most important limitations of our model are (a) the implicit assumption that electrons do not feel the atomic potential once they are excited to continuum states, (b) the ansatz for χ, and (c) the use of rate equations for the time evolution of the Auger state. All three assumptions can be justified for the parameters in the given experiment. It was verified by comparison with the complete numerical solution of the time–dependent Schrödinger equation that for the continuum with rather large energies of $\sim 40\,eV$ the impact of the atomic potential is indeed negligible [1]. By varying χ we verified that the exact shape of χ is unimportant, unless quantitatively accurate total electron yields are needed. The use of rate equations is justified, when only a single "gateway" Auger state is involved and when the characteristic time of its generation is short compared to its decay. Note that in the present experiment several Auger lines are involved, but the states are energetically well separated to justify, in first order, their treatment as independent states. A typical interest of future time–resolved attosecond experiments will be the investigation of more complex dynamical situations with several states involved. For such situations, our model must be further extended.

References

[1] M. Kitzler et al., Phys. Rev. Lett. **88**, 173904 (2002).