Energy Conservation in the Picosecond and Subpicosecond Photoelectric Effect

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The energy balance of the picosecond photoelectric is compared with that of the *subpicosecond* photoelectric effect. A full accounting of the energy must include the phase modulation (blue shift) induced on the laser beam by the ionization event itself.

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In a fascinating recent paper, Freeman *et al.*[1] published the kinetic-energy spectra of photoelectrons emitted from xenon atoms by multiphoton ionization. Conservation of energy implies that the electron kinetic energy is quantized in a multiphoton generalization of Einstein's famous photoelectric effect equation:

$$E=(n+s)\hbar\omega-I_0, \tag{1}$$

where E and $\hbar\omega$ are the electron kinetic energy and photon energy, respectively, and I_0 is the ionization potential of the atom. The integer n is the minimum number of photons required and integer s is the excess number of photons actually absorbed. For picosecond laser pulses, the electron kinetic energy has been observed to be beautifully quantized[2] in exactly the manner given by Eq. (1).

With *subpicosecond* pulses, Eq. (1) is disobeyed! The quantized electron peaks shift to energies which are *lower* than expected, and the distribution smears out and broadens at the shortest laser pulses. In this Letter I would like to answer: Where is the missing energy in the *subpicosecond* photoelectric effect? On the other hand, why does the picosecond photoelectric effect conserve energy in the normally expected way?

I believe that the answer to these questions revolves around the phase modulation induced on the laser beam by the ionization event itself. The ionization of a gas produces a definite change in its index of refraction. The optical polarizability of free electrons is much larger than and of opposite sign to the polarizability of neutral atoms. This sudden change of refractive index phase shifts a laser beam to higher frequencies. In a dense gas the ionization process can be accompanied by as much as a 5% or a 10% blue shift[3] in the laser frequency. The phase ϕ induced[4] by the ionization process is proportional to the free-electron polarizability

$$\phi=(2\pi e^2/m\omega c)N/A, \tag{2}$$

where e and m are the electron charge and mass, respectively, and N is the number of electrons per area A in the path of the laser beam on differentiation of Eq. (2), the phase shift can then be expressed as frequency shift $\Delta\omega$:

$$\Delta\omega=d\frac{\phi}{dt}=\frac{2\pi e^2}{m\omega c}\frac{dN}{A\,dt}, \tag{3}$$

where $dN/A\,dt$ is the rate of growth of ionization number per unit area. The energy transfer associated with this blue shift increases the energy of the photon field by $dn\,\hbar\Delta\omega$, where dn is the differential number of photons to pass through area A in a time dt. Therefore this differential energy transfer $d\Omega$ can be written as

$$d\Omega=\frac{2\pi e^2}{m\omega^2 c}\frac{\hbar\omega\,dn}{A\,dT}dN. \tag{4}$$

The expression $\hbar\omega\,dn/A\,dt$ is nothing more than the power flux of the electromagnetic field $cE^2/8\pi$, where E is the peak electric field and c the speed of light. Rewriting the differential energy transfer, we obtain

$$d\Omega=\frac{e^2E^2}{4m\omega^2}dN. \tag{5}$$

For an ionization number dN of unity, the energy transfer back to the laser beam during an ionization event is simply the famous[5] "quiver energy," $e^2E^2/4m\omega^2$, the oscillatory kinetic energy of an electron in an electromagnetic wave. By integration of Eq. (5) over a laser pulse, during which a single photoelectron is generated at t_0 which subsequently leaves the focal volume at t_1, the total energy transfer Ω becomes

$$\Omega=\left[\frac{e^2E^2(t)}{4m\omega^2}\right]_{t_1}^{t_0}, \tag{6}$$

where $E(t)$ is the laser electric field at time t during the pulse. This is the main result. We can now apply it to the physically different cases of picosecond and subpicosecond laser pulses.

In the picosecond case, free electrons are generated in the laser beam but they have time enough to leave the focal spot during the laser pulse. In the example given in Ref. 1, an electron with 2-eV kinetic energy can depart from the 20-μm focal spot in <20 psec. *The phase modulation during the ionization process at t_0 is exactly*

canceled by the reverse phase modulation at t_1 when the electrons leave the focal volume. From the beginning to the end of the laser pulse there is no net phase shift or net frequency shift. As a result of the rapid departure of the electron from the focal volume, Eq. (6) cancels. Energy balance does not require consideration of frequency modulation of the laser beam. The mean excess kinetic energy above the minimum required, $< s > \hbar\omega$, is mostly due to fact that the photoelectron is born not at rest, but in a coherent "quivering" state. Similarly, there can be additional excess energy due to a centrifugal barrier[6] for circularly polarized light. In all these cases electrons appear at the discrete kinetic energies given by Einstein's photoelectric Eq. (1).

For subpicosecond pulses the electrons will remain in the focal volume until the time t_1 when the laser electric field $|E(t)|$ has become smaller. Then Eq. (6) will become nonzero but still small. The initial effect is a shift of the discrete electron kinetic energies by a small fraction of the quiver energy. The electrons appear at energies lower than that given by Einstein's photoelectric equation. The likelihood of remaining behind in the focal volume until the end of the laser pulse increases toward unity for the shortest pulses. Then the full quiver energy will be transferred, tending to cancel the mean excess kinetic energy $< s > \hbar\omega$, washing out the discrete structure, and leaving little electron kinetic energy.

To summarize, when photoelectrons escape from the focal volume during a laser pulse, there is no net frequency modulation and they appear at discrete kinetic energies as given originally by Einstein. If they remain behind in the focal volume they cede their discrete quiver energies to a blue shift of the laser beam as a whole. The blue shift has actually been observed in the ionization of a dense gas[3] but is infinitesimally small when only a single photoelectron is involved.

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