Comment on "Energy Conservation in the Picosecond and Subpicosecond Photoelectric Effect"

In a recent Letter, Yablonovitch[1] pointed out that a full accounting of energy balance in the subpicosecond photoelectric effect must include the blueshift induced on the laser pulse by the ionization event itself. He showed that this extra energy acquired by photons not absorbed in the ionization process is mathematically equivalent to the "quiver energy" $e^2E^2/4m\omega^2$ of the freed electrons in the peak electric field E of the light pulse.

In our judgment, this Letter correctly describes the overall energy *balance* in the subpicosecond photoelectric effect, but ambiguously describes the energy *transfer* to the light field which produces the blueshift. This ambiguity occurs because the "ionization event" is a complex sequence consisting of the light-neutral-atom interaction during the leading edge of the pulse, generation of a free electron at time t_0, quivering of the free electron in the remaining part of the pulse, and passage of the trailing edge. When during this complex process does the blueshift occur? Yablonovitch is ambiguous on this point. Although his analysis identifies the blueshift with "the phase modulation during the ionization process at t_0" —i.e., the instant of generation of the free electron —he later ascribes the blueshift—incorrectly, in our judgement—to events in the trailing edge of the pulse: quivering electrons which "remain behind in the focal volume... cede their discrete quiver energies to a blue shift of the laser beam as a whole." The recently developed capability to observe blueshifts with femtosecond pulses[2] permits these two pictures of the blueshift dynamics to be distinguished experimentally. To support our argument we present a new femtosecond pump-probe measurement which shows unambiguously that the blueshift occurs at the expected instant of ionization t_0, which because of ionization saturation occurs *well in the leading edge* of a sufficiently intense pulse, not from a delayed transfer of electron quiver energy to the light field in the trailing edge of the pulse.

Figure 1(a) presents a calculation of the blueshift of a weak 100-fs, 620-nm probe pulse propagating collinearly with, and at varying time delays Δt within, a 100-fs, 620-nm pump pulse focused at $f/2$ to peak intensity 10^{16} W/cm^2 in 1-atm argon gas. At this intensity dN/dt, obtained by modeling ionization probability using Keldysh theory and then deriving ionic density growth from a rate-equation analysis, reaches maximum at $\Delta t = -60$ fs, a measurable time interval before the electron quiver energy peaks and subsides. The Drude model predicts that the probe blueshift tracks dN/dt in time, resulting in maximum blueshift also at $\Delta t = -60$ fs. Figure 1(b) shows the results of a corresponding experiment performed with our laser system[2] in which the probe pulse was separated from the orthogonally polarized pump after the interaction region by a polarization analyzer, and its spectrum recorded versus Δt by an optical multichannel analyzer. We determined $\Delta t = 0$ by slightly turning the pump polarization prior to the measurement, then varying Δt until fringes appeared in the transmitted beams from optical interference of pump and probe. By maximizing fringe contrast, $\Delta t = 0$ was determined to within ± 20 fs. Maximum blueshift is observed at $\Delta t = -60$ fs, in good agreement with the calculation. For $\Delta t \geq 0$, when the peak quiver energy is returning to the light field, no blueshift is observed. We conclude that the energy transfer responsible for the blueshift occurs instantaneously upon freeing of the electrons from the atom. The quiver energy of already freed electrons is reradiated later at the original (unshifted) light frequency, as in a linear dielectric response.

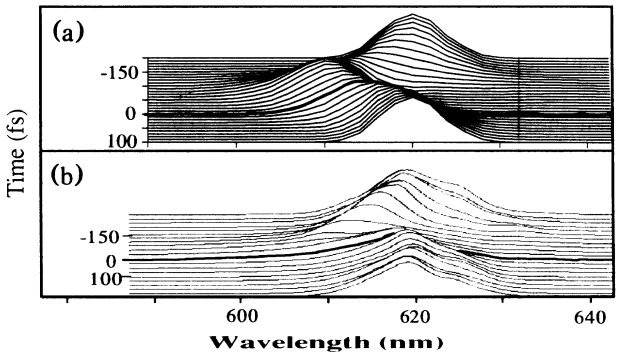

FIG. 1. (a) Calculation of blueshifted spectra of a 100-fs probe pulse at various time delays from the peak of a strong ionizing pulse focused to 10^{16} W/cm^2 in 1-atm argon. (b) Time-resolved measurement of probe blueshifts.

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