

PACS numbers: 06.60.Jn; 42.79.Pw; 41.85.–p
DOI: 10.1070/QE2013v043n04ABEH015120

Electron-optical metrology with femtosecond time resolution (theory and experiment)

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Abstract. **The principles of simultaneous spatial and temporal focusing of photoelectron beams in nonstationary electric fields are theoretically studied and experimentally realised as applied to time-analysing image-converter tubes (ICTs). It is theoretically shown that the use of nonstationary focusing electric fields makes it possible to surpass the theoretical time resolution limit of ICTs (10^{-4} s) determined for stationary fields by Zavoisky and Fanchenko in the 40s–50s of the last century. The possibility of forming electron packets with an energy of 10–30 KeV and a subfemtosecond duration (below 10^{-15} s) gives impetus to the development of time-resolved electron diffraction, which is a direct method of investigation of atomic-molecular dynamics in solid and gaseous media.**

Keywords: *time-analysing ICTs, photoelectron gun, time aberrations, nonstationary electric fields, temporal compression of photoelectron packets.*

1. Introduction

The two main advantages of accelerated electron beams – the rather short de Broglie wavelength ($\lambda \sim 10^{-2}$ nm at a beam energy of 20–30 keV) and the considerably (by 10^6 times) stronger (compared to photons) interaction with matter – make them rather effective for studying the fundamental properties of matter by time-resolved electron diffraction (TRED) [1–4]. The schematic diagram of a setup for TRED experiments is shown in Fig. 1.

A pulsed laser (*1*) emits a subpicosecond optical pulse, which is split into two pulses by a semitransparent mirror (*2*). The first pulse passes through the delay line (*3*) and excites the studied sample (*4*). The second pulse enters the electron gun (*5*) and, interacting with a photocathode (*6*), generates a photoelectron beam, which then is accelerated and focused on the sample by the electrode system (*7*). The electron beam interacts with the atomic-molecular system of the sample and forms a diffraction pattern on an image detector (*8*) at the instant of incidence of the exciting laser pulse on the sample. Varying the time interval between the instants of incidence of the initial laser pulse and the electron beam on the sample, one can receive direct information on transition processes inside the studied sample at the atomic-molecular level.

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Received 24 December 2012
Kvantovaya Elektronika **43** (4) 392–395 (2013)
Translated by M.N. Basieva

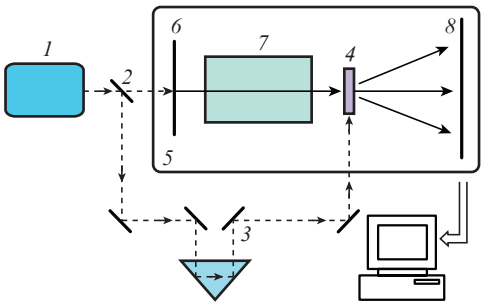

Figure 1. Schematic diagram of the setup for TRED experiments: (*1*) femtosecond laser; (*2*) semitransparent mirror; (*3*) optical delay line; (*4*) sample under study; (*5*) photoelectron gun; (*6*) photocathode; (*7*) electrode system; (*8*) image detector (for example, electron-sensitive CCD matrix).

2. Theoretical bases of focusing in stationary and quasi-stationary fields of photoelectron beams in ICTs

The TRED method time resolution in the transmission regime is determined mainly by the duration of the electron pulse propagating through the target. Modern self-mode-locked lasers emit optical pulses with a duration of several femtoseconds, but the formation of a photoelectron beam with a similar duration is a serious problem. In this work, we summarise the results achieved at the Photoelectronics Department, GPI, RAS during the last decade in the field of creation of time-analysing ICTs and photoelectron guns with subpicosecond resolution. The major obstacle in advancing the time resolution of ICTs to the femtosecond region is the spread of the initial photoelectron energies $\delta\varepsilon$, which is 0.3–1 eV for available classical photocathodes operating in the visible and near-IR regions. According to the Zavoisky–Fanchenko formula [5], the time spread of electrons in a beam formed in a static focusing field depends on their initial energies as

$$\delta T = \frac{\sqrt{2m}}{eE}\sqrt{\delta\varepsilon}\,, \qquad (1)$$

where m and e are the electron mass and charge and E is the electric field strength near the photocathode surface. It is important to note that the temporal chromatic aberration (1) cannot be in principle avoided in a static electric field and can be only decreased by increasing E. Using the theoretical electron optics terminology, we can assert that the first-order temporal focusing (ideal temporal focusing) is principally unachievable in static fields. From (1), it directly follows that, to achieve a temporal resolution better than 100 fs in static fields, it is necessary to increase the near-cathode field strength

E to ~25 kV mm^{-1} and higher. Taking into account restrictions related to the occurrence of an electric breakdown in vacuum, this brute force solution of the problem requires significant engineering efforts.

An efficient method of temporal focusing of photoelectron beams using nonstationary electric fields, which makes it possible to considerably surpass the limit dictated by the Zavoisky–Fanchenko formula for static fields, was proposed for the first time in [6] and then developed in [7–12]. The theoretical and computer calculations performed in these works showed that, in contrast to static electric fields, a properly chosen nonstationary field may allow one to completely eliminate the first-order temporal chromatic aberration and, hence, to realise the first-order temporal focusing. This is explained by the fact that a time-dependent electric field causes additional electron energy spread in the beam, so that the particles at the trailing edge of the beam begin to move faster than the particles at the beam front. After some time, the rear particles outrun the front particles, and the instant at which the electron beam duration becomes minimal determines the spatial position of the first-order temporal focus into which the sample under study must be placed. This principle to a certain extent is similar to the principle of grouping of charged particles in klystrons. However, a significant difference is that, to record a diffraction pattern in photoelectron guns of the considered type, one must not only ensure the temporal focusing of the electron beam on the sample, but also ensure its spatial focusing on the image detector.

Nonstationary electric fields make it possible to achieve first-order temporal focusing and considerably decrease the temporal broadening caused by Coulomb effects. This conclusion was confirmed by our detailed numerical experiments, in which the Coulomb interaction was modelled based on a specially designed general fractal algorithm [10].

In the first-order temporal focus, the contribution of the second-order temporal aberrations turns out to be considerable. If the initial laser pulse duration $\delta\tau$ is sufficiently high, then the pulse duration on the sample quadratically depends on $\delta\tau$. Shortening of the laser pulse can lead to a shortening of the electron pulse in the sample plane, but only to a limit determined by the existence of the second-order temporal chromatic aberration. As was shown in [6], this aberration can be completely eliminated by applying an additional magnetic field, which, in combination with increasing rate of change of the electric field, is a rather promising way to further shorten the electron pulse on the sample. However, even in the case of complete compensation of the second-order temporal chromatic aberration, the well-known Liouville theorem on the conservation of phase space volume claims that the existence of other (mixed) aberrations does not allow us to surpass a limit that, in fact, is an analogue of the Heisenberg uncertainty principle.

3. Computer simulation and practical realisation of a hybrid electron-optical device 'time-analysing ICT–electron gun'

To confirm the theoretical possibility of temporal focusing of photoelectron beams using nonstationary electric fields, the researches of the Photoelectronics Department, GPI, RAS calculated and fabricated an experimental photoelectron gun. Computer simulation was performed using ELIM/DYNAMICS [13] and MASIM [14] applied software packages designed by the researches of our Department.

One of the latest variants of hybrid electron-optical devices 'ICT–electron gun' developed and fabricated at the Photoelectronics Department, GPI, RAS is shown in Fig. 2. These hybrid devices differs from the previous ones by the photocathode–grid unit, which is designed in the form of lumped capacitance with a low-inductive system for applying pulsed voltage to the photocathode–grid gap. The modified hybrid device in the ICT regime must have a time resolution no worse than 100 fs at a pulsed field strength no lower than 25 kV mm^{-1} (electric pulses with an amplitude exceeding 12.5 kV are applied to the photocathode–grid gap 0.5 mm long). The numerical simulation shows that the same design in the photoelectron gun regime allows electron pulse compression to 35 fs.

Figure 2. Modified hybrid device 'ICT–photoelectron gun'.

4. Experimental results

The schematic diagram of the photoelectron gun is shown in Fig. 3. A laser pulse irradiates a narrow slit in the photocathode centre (*1*) and creates a photoelectron beam, which then is accelerated to an energy of 3 keV by a fine-structure grid (*2*) spaced from the photocathode by 1 mm. After passing through the fine-structure grid, the beam propagates through a region of a nonstationary electric field controlled by the electrode (*3*), to which a time-dependent voltage is applied. The rate of change of the pulsed voltage according to a particular temporal focusing regime is several kilovolts per nanosecond. The time-dependent electric field generator is strictly synchronised with the initiating laser, which provides the possibility of varying the time delay between the instants of the photoelectron beam emission from the photocathode and the beginning of generation of the electric field responsible for temporal focusing of the beam. As was mentioned above, in the course of the beam propagation in a nonstationary (in our case, linearly increasing) electric field, the energies of the frontal and rear electrons of the beam become different and, as is shown in Fig. 3, the aberration coefficient T_τ responsible for temporal focusing begins to decrease and turns to zero in the temporal focusing plane behind the anode (*6*). Electrodes (*4*) and (*5*) fulfil spatial focusing of the beam so that the spatial image plane coincides with the image detector (for example, with the plane of an electron-sensitive CCD matrix), while the

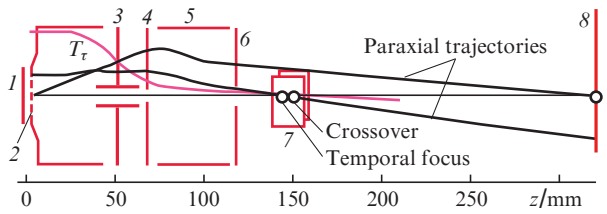

Figure 3. Schematic diagram of a photoelectron gun for spatiotemporal focusing of electron beams using stationary and nonstationary electric fields: (*1*) photocathode; (*2*) accelerating fine-structure grid; (*3*) electrode generating an electric field linearly changing in time; (*4*), (*5*) electrodes for spatial focusing of the beam; (*6*) anode; (*7*) dynamic deflector; (*8*) image detector.

beam crossover almost coincides with the temporal focus. A dynamic deflector (*7*) scans the beam over the screen with a phase velocity close to three speeds of light, which allows us to measure the electron pulse duration on the sample with a sufficient accuracy. Figure 4 shows the results of temporal focusing of photoelectron beams using photoelectron guns developed by us [15].

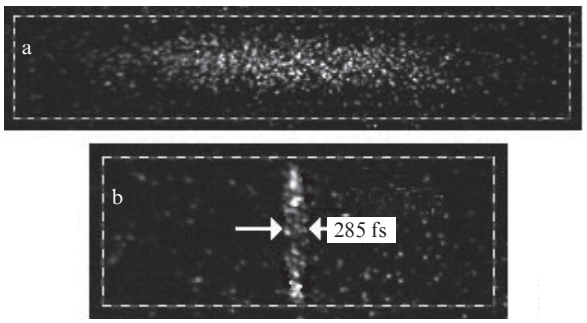

Figure 4. Chronographic electron beam image obtained using a 7-ps optical pulse of a semiconductor laser without temporal focusing in the slit scan regime (a) and chronographic image of a 285-fs electron beam obtained by temporal focusing (rate of change of the electric field used for temporal focusing ~2.4 kV ns^{-1}; scanning velocity ~2.8*c*, where *c* is the speed of light) (b).

To further shorten the initial photoelectron packets [16], it is necessary to increase the rate of change of the focusing electric field, which involves the creation of new pulse circuit technique. Moreover, it is necessary to search and probably develop a new generation of active elements (avalanche transistors, sharpeners, etc.) for commutation of 1–10 kV electric pulses with the transient response time not exceeding 100 ps. In other words, one must create circuit devices that can form kilovolt electric pulses with a rise rate no smaller than 10^{13} V s^{-1}. At the same time, the actuation time of these devices must not exceed a few nanoseconds with instability of several picoseconds. One of the approaches used by us to solve this problem consists in the development of pulsed systems containing chains of serially connected avalanche transistors and semiconductor sharpeners (Fig. 5).

5. Conclusions

The main results of the research performed at the Photoelectronics Department during the last decade can be formulated as follows.

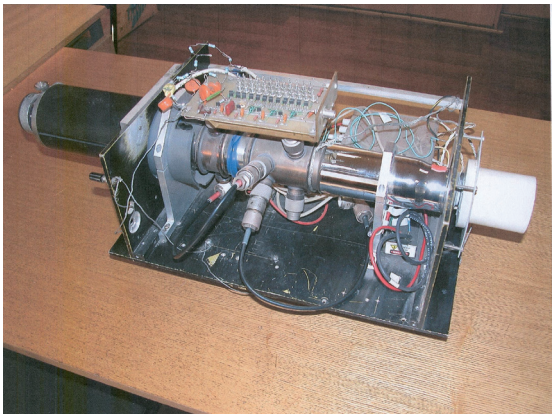

Figure 5. Experimental camera for static and dynamic tests of the hybrid device 'ICT–photoelectron gun'.

(1) A detailed aberration theory of spatiotemporal focusing of photoelectron beams in nonstationary electric fields is developed. It is shown that, in the properly chosen nonstationary field, it is possible to realise the first-order temporal focusing, which is impossible in static fields in principle. The Zavoisky–Fanchenko formula is directly generalised to the case of nonstationary fields, and it is shown that the use of these fields allows one to considerably (at least by an order of magnitude) exceed the theoretical time resolution limit for photoelectron beams (10 fs) determined by E.K. Zavoisky and S.D. Fanchenko for stationary focusing fields. It is shown for the first time that, in contrast to stationary fields, spatial charge effects in nonstationary electric fields can be to a great extent compensated by optimal choice of parameters of the electric field responsible for temporal focusing.

(2) Using computer simulation with applied software packages developed at the Photoelectronics Department, a series of experimental hybrid electron-optical devices 'ICT–photoelectron gun' was designed and fabricated. This photoelectron device is a unique electron-optical system which, for the first time in the history of electron-optical instrument-making, simultaneously provides both temporal and spatial focusing of photoelectron beams.

(3) A series of test experiments performed using the created hybrid device 'ICT–photoelectron gun' confirmed the theoretical statements and computer simulations, which formed the basis for practical realisation of this unique device. In particular, as a result of our experiment, a photoelectron beam obtained using a 7-ps optical pulse of a semiconductor laser was compressed to 285 fs. This experiment was performed at a rate of change of the time-focusing electric field of about 2.4 kV ns^{-1} and a phase velocity of beam scanning over the screen of ~2.8*c*, where *c* is the speed of light.

(4) It is theoretically shown that the experimental realisation of photoelectron packets with durations shorter than 100 fs requires nonstationary focusing fields with an amplitude of several kilovolts and a rise rate of no less than 10^{13} V s^{-1}.

Acknowledgements. The authors thank all the staff members of the Photoelectronics Department of GPI, RAS for their selfless work, which resulted in practical realisation of experimental samples of hybrid devices 'ICT–photoelectron gun'.

This work was supported by the state budget, by the programmes 'Femtosecond Optics and New Optical Materials'

and 'Extreme Light Fields and Their Applications' of the Presidium of the Russian Academy of Sciences, and by the International Science and Technology Centre (Project No. 2643).

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