Femtosecond electron gun for diffraction experiments

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ABSTRACT

The concept of an electron gun for generating pulses with a duration <10 fs at energies suitable for electron diffraction experiments is presented. The principle is based on an rf cavity oscillating in the TM_{010} mode. Laser pulses for photoemission are injected at a well-defined phase of the rf oscillation such that electrons with different initial velocities and different time delays arrive at a target within a very small temporal window. Coulomb broadening is prevented by reducing the number of electrons to the level of a single electron per pulse while increasing the repetition rate to the MHz range. The fs-electron pulses generated will advance the time-resolution of electron diffraction experiments to the level of a vibrational period of molecules.

Keywords: Electron gun, electron diffraction, rf-cavity, fs-pulses

1. INTRODUCTION

Time-resolved electron diffraction has become a powerful tool in the investigation of ultrafast processes in gases and solids [1-6]. In typical arrangements, ultrashort electron pulses are generated by illuminating a photocathode with a weak laser pulse and subsequently accelerating the electrons with a static voltage to energies of a few tens of keV. Such experiments allow investigation of transient processes in molecules in the gas phase with high temporal and spatial resolution.

However, in state of the art experiments the temporal resolution of transient electron diffraction is still limited to a few 100 fs. The reason for this is that electron pulses consisting of 10^3 - 10^4 electrons experience Coulombic repulsion caused by their space charge. This effect leads to severe broadening of the electron pulses upon propagation.

To eliminate the space charge broadening problem, it has been suggested that only a single electron per pulse be used and that the repetition rate be increased accordingly in order to get enough signal on the detector [7]. This calls for a laser with MHz repetition rates at energies high enough to generate the electron pulse and excite the molecules in a pump-probe experiment. Lasers suitable for implementing this idea have recently been developed at our laboratory [8]. Present parameters are a pulse energy of 500 nJ at a repetition rate of 2 MHz and a sub-40 fs pulse duration. Near-term improvements will scale the energy to the μJ level while reducing the pulse duration to <30 fs [9].

2. BREAKING THE LIMITS OF TIME RESOLUTION

In this paper we present the design of a photoinjector for generating electron pulses in the range of a few fs or even shorter. The basic idea is to apply the acceleration field at rf-frequencies rather than with a DC voltage. In this way two crucial advantages are obtained: a) The applicable field before breakdown can be considerably increased and b) by generating the electrons at an appropriate phase of the rf-cycle they can be made to arrive at a target within a few fs even with a significant initial velocity spread and with temporal delays.

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Millimeter-Wave and Terahertz Photonics, edited by Dieter Jäger, Andreas Stöhr,
Proc. of SPIE Vol. 6194, 61940J, (2006) · 0277-786X/06/$15 · doi: 10.1117/12.682975

Addressing the first of these points, we note that the duration of an electron pulse emitted by an accelerating gap is inversely proportional to the acceleration field. An empirical criterion for the breakdown field is given by the so-called Kilpatrick criterion. It is customary to write it in the form [10]

$$f = 1.643\, E^2 \exp(-8.5/E), \tag{1}$$

where f is the rf-frequency in MHz and E is the breakdown field in MV/m. By carefully designing the electrodes this limit can be exceeded by a factor of up to 5. Thus, the field given by eq. (1) constitutes an operating point rather than a limit. The breakdown field from the Kilpatrick criterion is displayed in fig. 1. The figure also shows the designation of various frequency bands, as traditional from radar technology. It is seen that already at frequencies in the UHF range the breakdown field is significantly enhanced. At X-band frequencies fields of up to 100 MV/m can be applied.

3. APPLYING AN RF-CAVITY FOR ELECTRON ACCELERATION

Standing wave rf-cavities are standard elements in accelerators (see, for example, [11]). In addition to a higher breakdown voltage they have the advantage of stable operation due to their narrow resonance. For acceleration to the subrelativistic energies needed in electron diffraction (30 - 100 keV) the design must be different from the standard accelerator design. Since velocities are only a fraction of the light velocity, different problems arise in synchronizing the particles with the cavity fields.

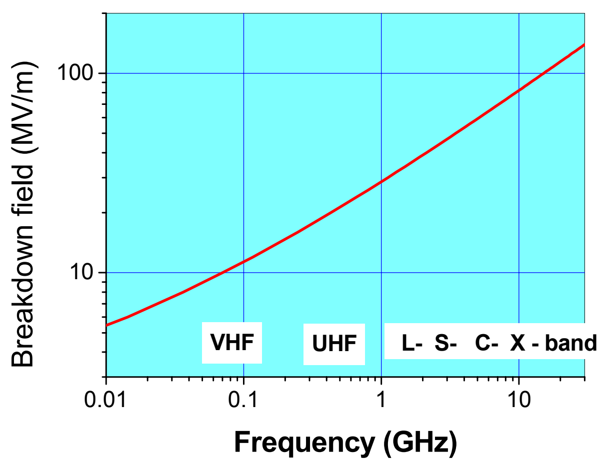

Figure 1: Vacuum breakdown field from Kilpatrick criterion and rf-frequency designations.

The essential feature used here to accelerate electrons to subrelativistic energies is the fact that the resonance frequency of a cylindrical cavity (a so-called pill-box) does not depend on its length but only on its diameter (see, for example, [11, 12]).Thus, one is relatively free in choosing an acceleration field and an applied frequency. It is only necessary to match the diameter of the pill-box to the applied frequency.

A pill-box cavity is shown in fig. 2 and the electric and magnetic fields associated with the lowest-order accelerating mode, the TM_{010} mode, are displayed. The fields of this mode are given by [11]

$$E_z = E_0 J_0\left(\frac{2.405}{r_0} r\right); \quad H_\Theta = H_0 J_1\left(\frac{2.405}{r_0} r\right); \quad H_0 = iE_0 / 377\,\Omega\,. \tag{2}$$

In these equations J_0 and J_1 are zeroth and first order Bessel functions and r_0 is the radius of the cavity. The relation between E_0 and H_0 applies in practical units. The resonance frequency of the cavity is given by

$$\nu_{010} = 1.147 \times 10^{10} / r_0 \qquad \text{(Hz)}, \tag{3}$$

with r_0 given in cm. All fields oscillate with frequency ν_{010}, the magnetic field with a phase of $\pi/2$ with respect to the electric one. The fields given by eq. (2) are optimal for particle acceleration: the electric field has a maximum on the axis, whereas the magnetic field on axis is zero. Thus deflection of the electrons by the magnetic field is precluded.

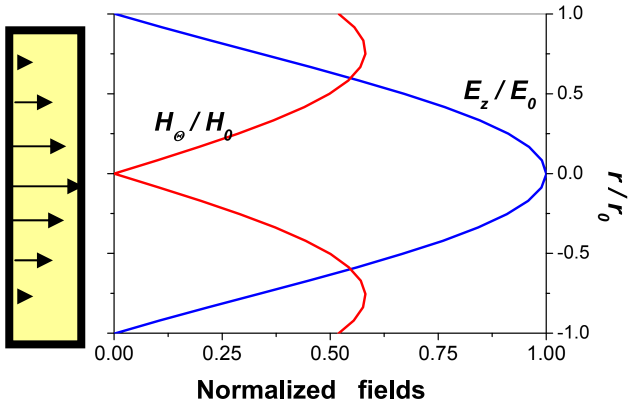

Figure 2: A pill-box with the electric fields schematically drawn into it (left) and the normalized values of the electric and magnetic fields as a function of radius (right). All fields oscillate without phase-shift along the cavity, the magnetic field being in quadrature with the electric one.

4. GENERATING FEW-FS ELECTRON PULSES

In principle, a fs electron gun with a pill-box cavity can be driven in the L- S- C- or X- bands by an accelerating field of some tens of MV/m (see fig. 1). It is advantageous, however, to use C-band frequencies (4-8 GHz) to reduce the size of the cavity and the required power (see below). It will be shown in the following that the maximum allowed breakdown field need not be applied. The photocathode is incorporated in the cavity at the rear side. The electrons leave the cavity through a small hole opposite the cathode.

The rapidly oscillating electric field in the pill-box cavity can be used to generate pulses in the few-fs regime. The basic idea is that electrons are generated at an rf-phase such that initially faster particles are accelerated slightly less to let the slower ones catch up at a target placed at a certain distance. We note that this method is applied to bunch compression in accelerators [13], but pulse durations in this case are in the ps-range. It will further be shown that particles emitted during a certain time window (due to the duration of the laser pulse) will also be temporally focused to <10 fs pulses at the target. In the following a fully relativistic one-dimensional analysis of electron motion in a pill-box and in the field-free region of the target (fig. 3) will be carried out. Consider a particle accelerated in a pill-box of length d propagating further to a target at a distance L from the end of the pill-box. Let the particle be launched at time zero into a field oscillating as $E_0 \cos(\omega t - \varphi)$, where ω is the resonant frequency of the pill-box cavity and a phase $\varphi > 0$ makes sure that the field is increasing while the particle propagates through the cavity.

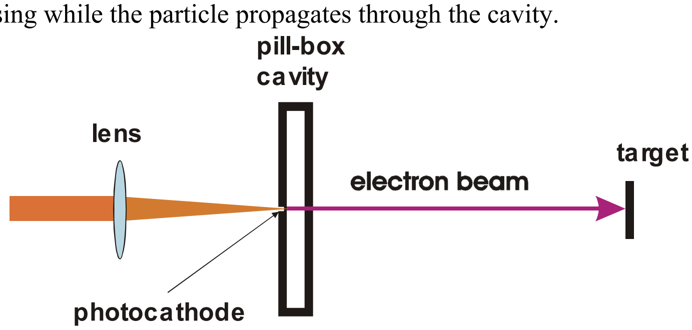

Figure 3: Geometry of pill-box cavity accelerator and target. The accelerating field is uniform in the propagation direction in the pill-box. The region from the end of the pill-box to the target is field-free. The photocathode is integrated into the cavity.

The equation of motion in the cavity will be given by

$$m\gamma^3 c\frac{d\beta}{dt} = eE\cos(\omega t - \varphi) \tag{4}$$

where m is the electron rest mass, β is the velocity divided by c and $\gamma = 1/(1-\beta^2)^{1/2}$ is the relativistic mass factor. Note the relativistic quantity $m\gamma^3$ on the left-hand side, which is sometimes called the "longitudinal mass".

Using the definition of γ, one can write eq. (4) as

$$\frac{d\beta}{\left(1-\beta^2\right)^{3/2}} = \frac{eE_0}{mc}\cos(\omega t - \varphi)\,dt\,, \tag{5}$$

which can be integrated to yield

$$\frac{\beta}{\left(1-\beta^2\right)^{1/2}} = \beta_q \sin(\omega t - \varphi) + C\,, \tag{6}$$

where $\beta_q = eE_0/(m\omega c)$ is the "quiver velocity" divided by c and C is an integration constant. Using the initial condition $\beta = \beta_i$ at $t = 0$, where β_i is the initial velocity of the electron divided by c, one obtains for the electron velocity at time t

$$\beta = \frac{f(t)}{\sqrt{1+f^2(t)}}\,, \tag{7}$$

where $f(t) = \beta_q\left[\sin(\omega t - \varphi) + \sin\varphi\right] + \beta_i\gamma_i$ and $\gamma_i = 1/(1 - \beta_i)^{1/2}$. Further integrating, one derives for the distance travelled by the electrons at time t

$$z = c\int_0^t \frac{f(t')}{\sqrt{1+f^2(t')}}\,dt'\,. \tag{8}$$

With this result one can finally calculate the time t_d required to traverse the cavity by solving for t_d the equation

$$d = c\int_0^{t_d} \frac{f(t')}{\sqrt{1+f^2(t')}}\,dt'\,. \tag{9}$$

The velocity of the electrons exiting the cavity becomes

$$\beta_d = \frac{f(t_d)}{\sqrt{1-f^2(t_d)}}\,. \tag{10}$$

One can now calculate the total time an electron propagates before reaching the target, as given by $t_{arr} = t_d + t_L$, where t_d is obtained from eq. (9) and $t_L = L/c\beta_d$ is the time of flight of the electron in the field-free region between cavity and target.

For optimum focusing in phase space the times t_{arr} for particles with different initial velocities should be as close as possible. Indeed, it turns out that for appropriate parameters arrival times t_{arr} are obtained in a range of less than 10 fs at initial velocities corresponding to up to 2 eV. This is illustrated in fig. 4, which displays the arrival times at the target relative to that of electrons with an initial velocity zero. Arrival times are displayed as a function of the initial electron energy with the phase as a parameter. The relevant parameters are 20 MV/m, an rf-frequency of 5 GHz, a cavity length of 2 mm and a distance to the target of 20 cm. Phase-focusing within about 5 fs is achieved at initial energies of up to 2 eV. A tolerable phase error would be ±0.002 rad, but a phase error of ±0.005 rad already increases the pulse duration to 30 fs. In these calculations a specific energy distribution of particles is not taken into account.

The foregoing analysis applies to electrons launched with different velocities at an infinitely short instant of time. To assess the effect of a finite pulse duration we make use of a theorem put forward by Monastyrskiy et al. [14], which states that to first order, effects of different velocities (chromatic aberration) and of different emission times (temporal aberration) are simultaneously corrected.

This remarkable result is verified by solving eqs. (9), (10) with small time shifts, to account for electrons released during a pulse of nonzero duration. The result is illustrated in fig. 5, which shows the times of arrival at the target as a function of time of emission of the electrons from the photocathode. It is seen that for electrons up to an initial energy of 1 eV a pulse duration of 50 fs (or equivalently, a delay in release time from the photocathode) still leads to an electron pulse < 10 fs. This result shows that - at least approximately - the effects of an initial velocity spread and of a nonzero pulse duration can be simultaneously corrected.

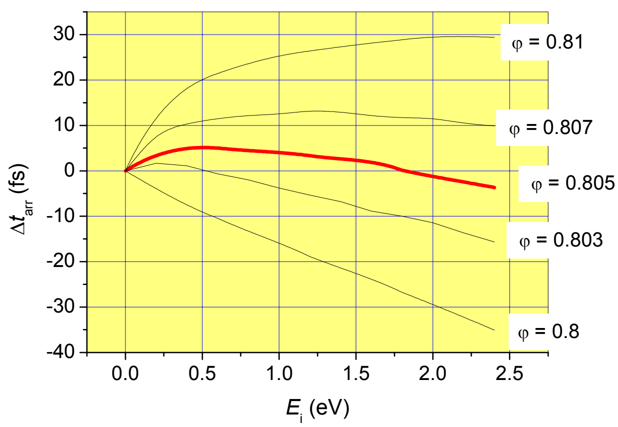

Figure 4: Times of arrival of electrons at a target 20 cm from the pill-box as a function of initial energy E_i for five different phases. Arrival time Δt_{arr} is given with respect to arrival time of electrons with zero inital velocity. For acceleration parameters see text. The optimum phase is $\varphi = 0.805$ rad.

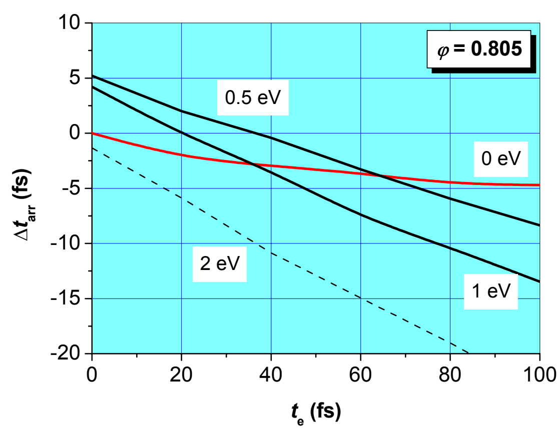

Figure 5: Times of arrival of electrons at a target as a function of time of emission t_e from a photocathode, with the initial electron energy E_i as a parameter. Arrival times Δt_{arr} are calculated with respect to the time of arrival of an electron released at time zero with zero velocity. The optimum phase from fig. 4 is chosen. Acceleration parameters are as in fig. 4.

It should not be overlooked that phase focusing results in a slight spreading of the energy spectrum of the electrons. Clearly, making electrons arrive simultaneously at a target is only possible if the electrons have slightly different velocities. This is demonstrated in fig. 6 which shows the energies of electrons arriving at the target as a function of initial energy with zero emission time. Fortunately, the generated energy spread is not significant: At the optimum phase ($\varphi = 0.805$ rad) the energy spread resulting from an initial energy of 2 eV is only 10 eV. Such an energy spread would not result in a considerable degradation of an electron diffraction pattern.

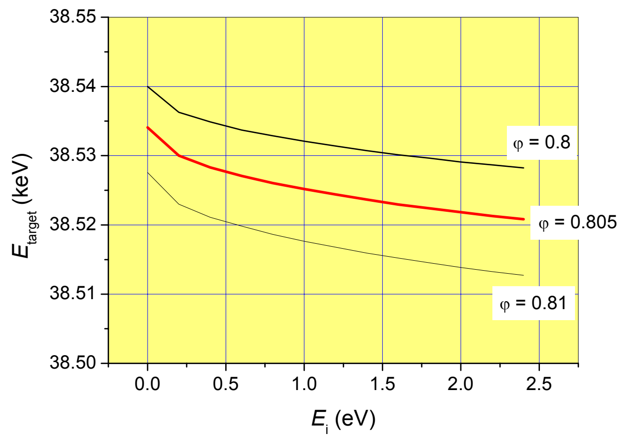

Figure 6: Energy on target of electrons accelerated by the pill-box cavity. The acceleration parameters are as in the previous figures.

5. SIMULATIONS

The pill-box cavity photoinjector was simulated using the GPT General Particle Tracer code [15]. For the simulations the acceleration parameters were chosen as in the above analytical calculations. In addition, the spot size of the emitting area was taken to be a circle with a FWHM of 20 μm and a Gaussian transverse distribution. The emission of the cathode was assumed to be isotropic with a temperature of 1 eV. The FWHM laser pulse duration was 10 fs with a Gaussian temporal shape. A sample of 1000 electrons was used to provide good enough statistics. However, space charge effects were neglected, in keeping with our proposition to use only a single electron per pulse. The phase of the electric field in the 2 mm long cavity was optimized to get the shortest pulse duration after 200 mm of free space propagation. This optimization yielded a phase $\varphi = 0.8034$ rad, slightly smaller than the analytical result.

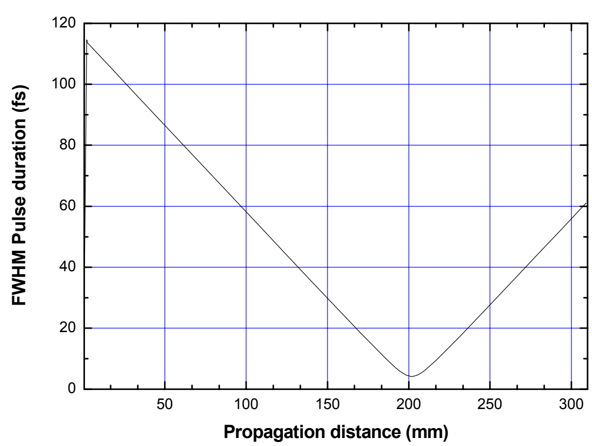

Figure 7: Simulated pulse duration vs. propagation distance. The acceleration parameters are as for fig. 4. The phase is optimized to $\varphi = 0.8034$ rad. Space charge is not taken into account.

The FWHM bunch duration is plotted in fig. 7. The pulse elongates to 110 fs at the cavity exit and is then compressed to a minimum duration of 4.2 fs after 200 mm of propagation. In keeping with the results shown in fig. 5, runs with a longer laser pulse duration also yielded an electron pulse duration <10 fs. The calculated normalized transverse emittance was 6 x 10^{-3} mm mrad. Preliminary runs with the space charge included show that with only 10 electrons the pulse is already significantly broadened to about 20 fs. A detailed account of the effect of space charge, using GPT with the space charge model as described in [16] will be presented in a forthcoming publication.

6. CONSIDERATIONS OF TECHNICAL FEASIBILITY

A number of considerations are called for to ascertain the technical feasibility of the pill-box accelerator. These concern ways to incorporate the photocathode into the pill-box, synchronization of the rf-frequency to the laser repetition rate, and the power dissipated to excite the rf-cavity. In the following, basic ideas addressing these problems are presented.

A possible design for a photocathode consists of a thin fused silica disc coated with a suitable photocathode material, e.g. copper. The coating must be thinner than the skin depth of the laser to avoid electrons being emitted with too high a temporal delay. The photocatode is situated at the center of the cavity, opposite to the exit hole of the electrons. Note that the power dissipated on the cathode is very small due to the low value of the magnetic field at the center of the cavity (see fig. 2). In principle the photocathode can be operated with ω, 2ω or 3ω - light from a titanium-sapphire laser using three, two or one photon, respectively, for photoemission. Experiments with <100 fs titanium sapphire pulses show, however, that single-photon photoemission is most favourable for generating a low initial electron temperature [17]. With third-harmonic light from a titanium-sapphire laser ($\lambda =$ 267 nm) at an intensity of 10^4 W/cm^2 a Cu-cathode yields an initial electron temperature of about 0.3 eV.

To achieve optimum phase focusing the rf-generator and the laser must be well synchronized. This can be achieved by a series of frequency dividers. For example, if the frequency ratio between the two is 1024, a series of 10 frequency-doublers would generate the required synchronization. The phase-must be controlled with a high but not unrealistic accuracy: As fig. 4 shows, a phase error of ±0.002 would still be tolerable, resulting in a timing-error $\Delta t = \Delta\varphi/\omega$ of ±64 fs. We note that at lower rf frequencies the synchronization condition is considerably relaxed. However, the rf-power required scales as $\nu^{-3/2}$ (see below).

An issue of concern is the rf-power required to sustain the cavity oscillation. Note that beam loading will be negligible, since the current generated is very low, and, moreover, only a small fraction of the cycles will draw a current in the cavity. Losses will therefore be entirely determined by surface currents along the walls of the cavity. The dissipated power by surface currents is given by [11]

$$P_d = \frac{R_{surf}}{2}\int_S |H|^2 ds\,, \tag{11}$$

where R_{surf} is the surface resistance of the material, H is the magnetic field at the wall, and the integral is to be taken over the cavity surface S. For a short cavity the contribution from the perimeter can be neglected and the surface integral is calculated

with H_Θ given by eq. (3). Using $\int_0^{r_0} rJ_1^2(kr)\,dr = \frac{r_0^2}{2}J_1^2(kr_0) \approx r_0^2/8$, we obtain for the dissipated power the simple expression

$$P_\mathrm{d} = 2.76 \times 10^{-6}\, R_{surf}E_0^2\, r_0^2\ (\mathrm{Watt}). \tag{12}$$

In this equation E_0 is in V/m, r_0 is in m and the prefactor has the dimension (Ohm^{-2}).

Consider a copper cavity with the parameters of the example in section 4, i.e. an accelerating field of E_0 = 20 MV/m, a frequency of 5 GHz, corresponding to an r_0 of 2.29 cm. The cavity length of 2 mm justifies neglecting the contribution of the perimeter. The surface resistance of copper is given by $R_{surf} = 2.61 \times 10^{-7}\ \omega^{1/2}$ Ohms [11], which becomes 4.6×10^{-2} Ohms at 5 GHz. Inserting these values in eq. (12), one obtains for the dissipated power $P_d = 27$ kW. Note that the proportionality $P_d \propto R_{surf}\, r_0^2$ results in a scaling of $P_d \propto \nu^{-3/2}$.

While a value of 27 kW is feasible for cw operation using high-power klystrons, it can still be reduced by cooling the cavity. At liquid-nitrogen temperature the surface resistance of copper is 1/6 of that at room temperature [18], resulting in a dissipated power P_d of only 4.5 kW. Further reduction would be possible for a superconducting cavity. The surface resistance for a superconductor is proportional to the square of the frequency and depends exponentially on the ratio T/T_c , where T is the temperature and T_c is the critical temperature [18]. For niobium at $T/T_c > 2.5$ ($T < 4$ K) one has $R_{surf} < 1$ μOhm and the dissipated power would go down to the 10 W level.

7. CONCLUSIONS

We have shown that an electron gun for sub-relativistic fs-electron pulses can be designed by using a cylindrical cavity excited by a resonant rf-field. This design has two main advantages: first the accelerating field can be increased without being limited by vacuum breakdown and, secondly, by choosing the appropriate phase of injection of the electrons the pulse can be shortened to <10 fs in spite of different initial electron velocities and injection times. The shortened pulse has to be payed for by a small energy spread of the electrons as they arrive at a target. Important technical issues, such as an appropriate photocathode design and the power required to excite the cavity oscillation, are considered and prove the feasibility of this concept. Such electron pulses will be an essential part of electron diffraction exeriments capable of exploiting the full capability of this technique.

ACKNOWLEDGEMENT

We thank the authors of the GPT code, Dr. S.B. van der Geer and Dr. M.J. de Loos, for fruitful discussions.

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