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Direct observation of space charge dynamics by picosecond low-energy electron scattering

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Abstract – The transient electric field governing the dynamics of space charge is investigated by time- and energy-resolved low-energy electron scattering. The space charge above a copper target is produced by high-intensity femtosecond laser pulses. The pump-probe experiment has a measured temporal resolution of better than 35 ps at 55 eV probe electron energy. The probe electron acceleration due to space charge is reproduced within a 3-dimensional non-relativistic model, which determines an effective number of electrons in the space charge cloud and its initial diameter. Comparison of the simulations with the experiments indicates a Coulomb explosion, which is consistent with transients in the order of 1 ns, the terminal kinetic energy of the cloud and the thermoemission currents predicted by the Richardson-Dushman formula.

Introduction. – The current which can be drawn from a hot cathode follows the Richardson-Dushman formula, where the current density is given by the filament temperature and the work function [1]. In such a quasi-static situation the electric fields due to the space charge of the emitted electrons decrease the thermoemission [2]. In pulsed electron sources, as *e.g.* needed for free electron lasers [3,4] or field emitters [5], laser pulses may be used to heat and emit electrons. In this case the field of the space charge has a transient behaviour and will, depending on its timing, also accelerate electrons [6]. There are a wealth of experiments that address the heating of an electron gas in a metal with laser pulses [7–11] and the concomitant space charge problem [12]. In these experiments the system is excited with an optical pump pulse and its evolution is probed after a given delay with another optical pulse, where emitted light [9] or electrons are used to probe the electron dynamics inside and outside of the solid. So far there are no experiments that report measurements of the time evolution of the electric fields due to emitted electrons.

Here we report electron scattering experiments where picosecond pulses of low-energy electrons were used to probe the dynamics of femtosecond laser pulse induced space charge. It is a significant extension of light-electron pump-probe experiments, which have, so far only been conducted with keV electrons [13–17]. The use of low-energy electrons has obvious advantages due to higher electron scattering cross-sections and a better absolute electron energy resolution, which then allows to time resolve the energy of backscattered electrons on the meV scale.

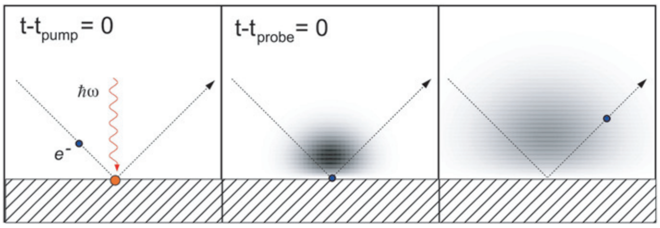

Fig. 1: (Color online) Schematic view of the space charge experiment for 3 time frames. At $t - t_{pump} = 0$ a laser pump pulse $\hbar\omega$ hits the surface. At $t - t_{probe} = 0$ the probe electron scatters on the surface. The shown probe electron e^- has a positive delay $t_D = t_{probe} - t_{pump} > 0$. The pump pulse excites the electrons in the solid, where a small portion of the hot electron gas may escape into the vacuum. The transient field of the electron cloud depends on the pump-probe delay and acts on the probing electrons.

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After improving the design of our pulsed low-energy electron gun [18] in terms of fluence by 3 orders of magnitude the evolution of space charge could be studied in recording the energy gain of 55 eV electrons specularly scattered off a Cu(111) surface. Figure 1 shows the scheme of the experiment: space charge is created by an intense laser pulse on a surface, where the evolving electron cloud is probed with a pulsed electron beam. At laser power densities in the GW/cm^2 range, the emitted charge is self-accelerated due to a Coulomb explosion [19] and then expands with superthermal energies into the hemisphere above the surface. Our experiment complements and confirms this picture with time-resolved measurements of the transient electrostatic potential of an expanding space charge cloud.

Experimental setup. – The experiments were carried out in a stainless-steel Ultra-High-Vacuum (UHV) system without shielding the earth magnetic field. All measurements were performed at room temperature at a base pressure in the 10^{-11} mbar range. The laser system consists of a commercial Coherent MIRA Ti : sapphire oscillator that emits pulses centered around the wavelength $\lambda_0 = 800$ nm with a spectral width $\Delta\lambda \sim 28$ nm and a time width Δt of about 55 fs. The output pulses can be amplified to higher pulse energies by a chirped-pulse Regenerative Amplifier (Coherent RegA 9050); after the amplification process, the pulse energy is $\sim 5\,\mu$J/pulse at a repetition rate of 250 kHz. The 800 nm laser light is split by a beamsplitter into two beams, one of which is directly sent towards the vacuum chamber, while the other is frequency doubled. The 400 nm beam passes through a delay stage, which can vary its optical path up to 60 cm and is then used to produce the electron pulses on a back-illuminated gold cathode of a home-built electron gun. The electron gun has a design with a time resolution below 5 ps. Separate experiments show an electron yield of ≈ 1 electron/pulse at a pulse energy of 1 nJ and a time resolution better than 35 ps at 55 eV primary energy. Above 1 nJ pulse energy electron pulse spreading due to space-charge effects in the electron gun occurs [20]. The measured time resolution is the fastest transient time in an electron beam blocking experiment [21]. It is likely to be limited by the space-charge dynamics in the pinhole of the beam blocking experiment, but not by the electron gun.

The space-charge dynamics above a Cu(111) surface were studied in a setup where the specular electron beam is backscattered and detected (see fig. 1). The Cu(111) surface may be easily prepared by standard cleaning procedures, and remains clean during the experimental scans that lasted up to 10 hours.

The probe electron energy of 55 eV was optimized for reflectivity and low spectral overlap with the cloud electrons. The scattered electrons were energy analyzed with a 100 mm mean radius hemispherical electron analyzer (Clam2). The pump beam is chopped with a frequency of about 20 Hz, and for each delay stage position the data are recorded with and without the pump beam, which allows to check the stability of the electron gun. The temporal coincidence (delay zero) and spatial overlap between the light pump and the electron probe is determined with the electron-photon correlator presented in ref. [21].

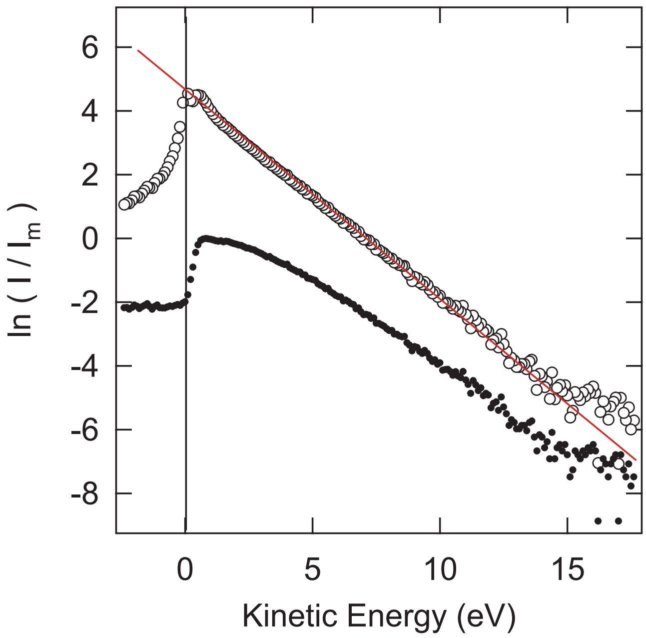

Fig. 2: (Color online) Logarithmic spectrum of electrons emitted by 800 nm, 10 mJ/cm^2, 100 fs laser pulses hitting a Cu(111) surface. If the spectrum (black dots) is normalized with $\sqrt{E_{kin}}$, a Boltzmann-type distribution with an effective energy $k_B T_c$ of 1.6 eV is found (open circles). I_m is the measured maximum intensity.

Results. – First, the electron energy distribution of the space charge that leaves the sample was measured. In fig. 2 the electron energy spectrum $I(E)$ of a space charge cloud from Cu(111) is shown on a logarithmic scale for focused laser pulses of $5\,\mu$J. With a laser focus of 0.05 mm^2 this yields a fluence of about 10 mJ/cm^2 or a power density of 100 GW/cm^2, which corresponds at 250 kHz repetition rate to a sample current of about 1 nA. The spectrum is measured at normal emission with a bias voltage of -50 V applied to the sample. The concomitant extraction field of 1 kV/m is not expected to change the shape of the energy distribution of the space charge electrons [19]. If the measured electron distribution is normalized with the escape probability of the electrons, we find an almost perfect exponential distribution.

The escape probability is given by electron refraction at the inner potential and is proportional to the momentum of the electrons in the vacuum, *i.e.* the square root of their kinetic energy [1,22,23]. The slope of the $\ln(I/\sqrt{E_{kin}})$ *vs.* E_{kin} curve translates in an average energy $k_B T_c$ of the cloud electrons of 1.6 eV. In a Boltzmann picture this energy correspoonds to a “cloud temperature” T_c of $2 \cdot 10^4$ K. The Richardson-Dushman equation

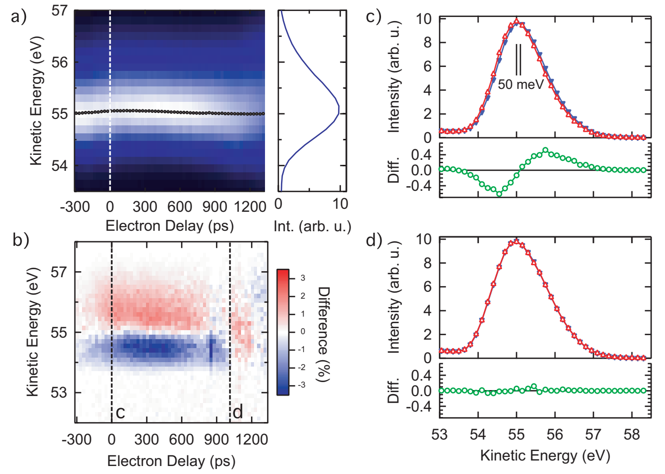

Fig. 3: (Color online) Spectra of specularly scattered electrons with 55 eV kinetic energy off Cu(111) in the "presence" of pump laser excitation with a pulse energy of 5 μJ. (a) Specular beam intensity with pump beam as a function of the delay $t_{probe} - t_{pump}$ between pump and probe pulse. The black open circles follow the peak maximum, where a time-dependent shift is noticed. On the right-hand side the energy spectrum sliced out at the 0-delay (marked with the dashed line) is shown. (b) Difference of the electron energy distribution $I_w - I_0$, where I_w is the intensity of the scattered electrons with pump and I_0 that without pump beam. (c) Energy spectra with (blue) and without (red) pump at coincidence (dashed line in (b)) labeled "c"). Note the shift of about 50 meV in the presence of the pump pulse. The bottom panel shows the difference between the two curves. (d) Energy spectra with (blue) and without (red) pump about 1 ns off coincidence (dashed line in (b)), labeled "d").

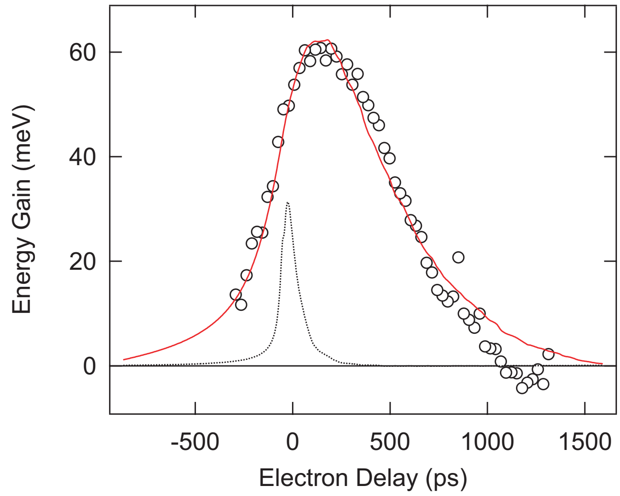

Fig. 4: (Color online) Kinetic energy gain of probe electrons ($E_{kin} = 55$ eV) in the presence of a space charge cloud *vs.* electron delay. Positive delays mean that the pump pulse hits the surface before the probe. A maximum shift of 60 meV occurs about 100 ps after coincidence. The red solid line is the best fit to the model with an energy distribution, as shown in fig. 2, with $6.3 \cdot 10^4$ electrons and an initial cloud diameter of 1.8 mm. The dotted line is the result for 5000 electrons in a cloud with 0.4 mm initial diameter.

predicts a current density j of thermoemitted electrons of

$$j = AT^2 \exp(-\Phi/k_B T) \tag{1}$$

with $A = 1.2 \cdot 10^6$ A/K^{2}m^2. This leads, with a thermal energy of $k_B T = 1.6$ eV, a work function Φ of 4.94 eV, a focus size of 0.05 mm^2, a pulse duration of 100 fs and a repetition rate of 250 kHz, a sample current of about 25 mA, which is way beyond the measured values. The large kinetic energy in the cloud is also at some contrast to measured electron temperatures in the solid after femtosecond laser heating which is in the order of a few 10^3 K [24]. It is, thus, an indication that the emitted electrons undergo a kind of self-acceleration in the course of their way to the detector, where the energy for the self-acceleration stems from the Coulomb energy in the cloud. This picture is substantiated by the measurement of acceleration of probing electrons, as will be shown in the following.

Figure 3 shows the results of the pump-probe experiment from the Cu(111) target. Pump pulses with a duration of 100 fs, a wavelength of 800 nm and a pulse energy of 5 μJ and probe pulses with 55 eV electrons were used. Figure 3(a) displays the electron beam intensity as a function of the delay between pump and probe, where negative delays mean that the probe electrons are hitting the sample surface before the pump pulses. In order to highlight transient changes, the difference $I_w - I_0$ is shown in fig. 3(b). I_w is the intensity of the elastically scattered electrons when the pump light is on the sample and I_0 the one without pump light. The energy spectrum of the probe electrons is affected by the pump pulse: we observe a delay-dependent energy shift towards higher kinetic energies. In fig. 3(c), at delay zero, this shift amounts to 50 meV, while after 1 ns delay the probing electrons are not accelerated anymore (fig. 3(d)). This allows to estimate the order of magnitude of the average accelerating field. From the duration of the transient of 1 ns and the energy gain of 55 eV electrons we find an average accelerating field of about 10 V/m, which corresponds to a surface charge density in a plate capacitor of about $10^9 e^-/\text{m}^2$.

Discussion. – In order to get a more quantitative picture, the energy shifts of the probing electrons shown in fig. 3(a) were determined for all delays by fitting Gaussians to the energy distribution curves. Figure 4 shows the corresponding peak shift of the electron spectra with pump light relative to that without pump light. The data peak after delay zero and the transient is non-Gaussian, *i.e.* has a slower decay compared to the rise at negative delays. It has to be noted that in contrast to light probes the rise at negative delays, *i.e.* when the probe electrons hit the surface before the pump pulse, is not due to the time resolution of the experiment. It merely shows that the field of the expanding space charge behind, also reaches the probe electrons.

The observed probe electron acceleration due to space charge is simulated by a non-relativistic model. This is justified since all relevant electron velocities were much smaller than the speed of light. The model is 3-dimensional but neglects the acceleration of the cloud. The initial cloud is simulated as a homogeneously charged disk of electrons which starts to expand from the surface at a time $t - t_{pump} = 0$. According to the empirical result shown in fig. 2, the electron energy distribution is proportional to $\sqrt{E_{kin}}\exp(-E_{kin}/k_B T_c)$, where the energy $k_B T_c = 1.6\,\text{eV}$ is taken from the measured kinetic energy distribution. The angular distribution of the electron trajectories is set to be proportional to $\cos(\theta)$, where θ is the polar emission angle as measured from the surface normal [23]. The initial cloud diameter and the probing electron spot were left as free parameters. The resulting electric field of the space charge cloud and its image along the probe electron trajectory is determined in summing the contributions of idividual cloud-electron trajectories, where the initial conditions were determined by a Monte Carlo algorithm that satisfies the above described model parameters. The integration of this field along the probe trajectory delivers the kinetic energy gain of the probe electrons as a function of the delay $t_D = t_{probe} - t_{pump}$. A comparison of the simulation with the experiment identifies an effective number of electrons in the space charge cloud. For the data shown in fig. 4 it is found to be $6.3 \pm 0.2 \cdot 10^4$ electrons in an initial disk with a diameter of 1.8 mm and a probe spot with a diameter smaller than 0.6 mm. In fig. 4 we also show for comparison a simulation for a cloud with 400 μm initial diameter and a point-like probe with 5000 electrons, which gives a limit of the shortest possible acceleration transients of 100 ps. The resulting cloud currents from the model without acceleration were compatible with the measured emission currents, though they are incompatible with the Richardson-Dushman formula (eq. (1)) using an emission temperature of $2 \cdot 10^4$ K. Also, the diameter of the initial cloud appears too large, compared to the laser focus, which is in the order of 0.05 mm^2. We take these deficiencies as an indication that the model assumption of no acceleration of the cloud is *not* correct, *i.e.* as a clue for a Coulomb explosion, which accelerates the electrons during emission. It is beyond the scope of this paper to model the explosion in detail, also because more experiments with different probe electron energies should provide a larger data base for the test of the models. Such a model would then also allow to determine the timescale on which the image charge evolves. However, we may reconcile the observed number of electrons in the cloud with the Richardson-Dunshman formula, if we consider electron temperatures after laser heating of ≈ 3500 K [24]. The terminal temperature of the cloud is explained if the initial Coulomb energy is transformed into kinetic energy. Indeed, a disk with a diameter of a laser focus of 0.25 mm and about $6 \cdot 10^4$ electrons has an average potential energy of 1.5 eV per electron and further confirms the picture of Coulomb heating in the early stage of space charge evolution.

Conclusions. – In conclusion it is shown that a time- and energy-resolved low-energy electron scattering experiment gives new and complementary insight into the dynamics of an expanding space charge cloud. The measured cloud energy and the transients in the order of 1 ns indicate that a Coulomb explosion self-accelerates a space charge cloud which is generated by μJ femtosecond laser pulses. If this time-resolved electron scattering experiment is expanded to diffraction of low-energy electrons (LEED) it will also become useful for the recording of structural changes on surfaces at ultra-short time scales.

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