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# The pulsed arc cluster ion source (PACIS)

**H.R. Siekmann, Ch. Lüder, J. Faehrmann, H.O. Lutz, and K.H. Meiwes-Broer**

Fakultät für Physik, Universität Bielefeld, W-4800 Bielefeld 1, Federal Republic of Germany

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**Abstract.** A new cluster source, the "PACIS", has recently been developed [1, 2]: a pulsed high-current arc is fired between two electrically isolated electrodes. In a stream of carrier gas, the nascent metal plasma cools down, thus undergoes significant aggregation. After supersonic expansion into high vacuum, the resulting cluster ions will be subject to investigation. Here, we present the current state of the source development and further technical details. The integral intensity is estimated by film thickness measurements and yields a deposition rate of, e.g., up to 2 Å per shot for lead. About 10% of the emitted material turns out to be charged. Time-of-flight measurements show similar cluster ion mass spectra as they are known from laser vaporization. Already single shot mass spectra display complete *n*-series which can be followed to $n \approx 40$. For heavy clusters, a significant velocity slip is estimated from the beam velocities.

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## I. Introduction

Spectroscopic investigation of clusters makes it necessary to produce the species in defined (ground) states [3]. For high melting point materials the production of "cold" clusters was not possible until Bondybey et al. [4] and Dietz et al. [5] introduced the concept of laser vaporization (of nearly any solid material) and subsequent cluster growth in a stream of a cold carrier gas. This technique has since become a reliable tool to produce intense cold cluster ion and anion beams without any additional ionizing agent [6]. Nevertheless, there is still the need to have a laser to excite the source, i.e., discharge energy is transformed into light, which carries the energy into the source and produces a plasma.

Only very recently, we reported on a method to circumvent the energy- (and money-) consuming laser light production, i.e., the pulsed high-current discharge is not fired inside the laser but directly in the source. In contrast to a similar setup used for the production of aerosol particles [7], for the growth of, e.g., pure metal or carbon cluster ions a high-power electric switch is necessary.

The metal cluster ion mass spectra of this "pulsed arc cluster ion source" (PACIS) turn out to be similar to those obtained with a laser vaporization source. In addition, the source is suited to create metal doped rare gas cluster ions, e.g., $Ar_nAl^+$ [1]. So far such mixed clusters have been generated by laser vaporization [8]. As a further example of the large scope of this new source, Busmann et al. used ethylene as a carrier gas to produce intense $C_n$ and $C_nH_m$ cluster ion beams [9].

In this communication we will give more detailed information about the source and present intensity data as well as typical mass spectra.

## II. Experimental

The vacuum apparatus employed in the present experiment is similar to that used in our group to create metal clusters via the laser vaporization method and to investigate them by photoelectron spectroscopy [6, 10]. In short, metal plasma bunches, created in the pulsed arc discharge, are seeded by a gas and expand freely into a 200 l vacuum chamber. A 2000 l/s oil diffusion pump, backed by a roots blower, evacuates the source chamber. Positively as well as negatively charged cluster ions drift in the supersonic jet through the skimmer into the two-stage acceleration device of the time-of-flight mass spectrometer (TOFMS); here they experience pulsed acceleration. A set of einzel lenses guides the cluster ion beam over 1.4 m on a channelplate detector. The signals are recorded by a 350 MHz waveform digitizer with fast adding memory (Le Croy 9450).

On its way to the detector the beam passes a small magnetic bottle TOF electron spectrometer with a 10 cm flight tube. It may be used to investigate photoelectrons for identification of high-mass clusters which cannot be

registered with the channelplates. Details of this "photodetachment detector" will be published elsewhere.

The basic idea of the "PACIS" has been described briefly in earlier publications [1, 2] and will be discussed below in more detail. First we will comment on the two different power supplies used, and then will give a description of the source construction and operation.

Two different versions of the discharge circuit have been tested, both of which are suited to produce cluster ion and anion beams with at least the intensity of the laser vaporization source. The first discharge circuit stems from an excimer laser (Lambda Physik, EMG 102). A grounded grid thyratron serves as high energy switch and yields a maximum of $3 \cdot 10^{-3}$ Coulomb/shot at a repetition rate of 100 Hz; the applied discharge voltage varies from 2 . . . 12 kV.

In order to increase the charge load per shot and thus the amount of material vaporized, a second power supply has been developed. It consists of a home-made "high-power switch" which is fed by a DC supply with adjustable voltage up to 1300 V, see Fig. 1a. A monoflop network triggers a charging as well as a discharging thyristor. The source discharge current is limited by a resistor to below 1000 A; up to 30 mC/shot are supplied by the capacitor C1. The electrical components have been chosen for a repetition rate of up to 100 Hz.

The operation at maximum power and repetition rate requires source cooling, since otherwise 100°C may be exceeded easily. When using $LN_2$-cooling cluster aggregation [11] is enhanced, whereas contaminations by, e.g., oxygen decreases. Furthermore, the pulse to pulse stability increases.

The design of the PACIS is shown schematically in Fig. 1b. An insulator block made of boron nitride or Macor (B) is fixed and supported by two aluminium plates (A) and contains two cylindrical electrodes with a diameter of 5 mm and a spacing of 1–2 mm. A pulsed valve (General Valve, Ser. 9) supplies pure or mixed seeding gas (He, Ar, . . .) through a 1 mm diam by 10 mm long channel. The discharge is fired inside the 0.5–2 ms long carrier gas pulse, the resulting plasma being flushed through a 2 mm diam 10 mm long channel into the subsequent "mixing chamber" (C). This has a volume of about 100–400 $mm^3$ and serves as thermalization zone while effectively mixing the hot helium/plasma part with colder portions of the carrier gas [12]. The resulting mixture is again compressed to a 1.5 mm diam 5 mm long channel followed by a 30 mm long 15° total angle cone (D).

The "mixing chamber" (C) turns out to be quite important when the source is operated under high-power conditions, i.e., with the thyristor driven power supply. It should be removed, however, when using low power as is necessary for, e.g., the creation of metal-doped rare-gas clusters, or when using the thyratron power supply. This seems reasonable as the latter yields discharge times of about 100 ns, compared to more than 10 µs with correspondingly higher amount of energy provided by the thyristor-based power supply.

Apart from using the mass flow rate to calculate the pressure in the source [13], which yields 1.5 bar, this pressure can be estimated in an unconventional way: The

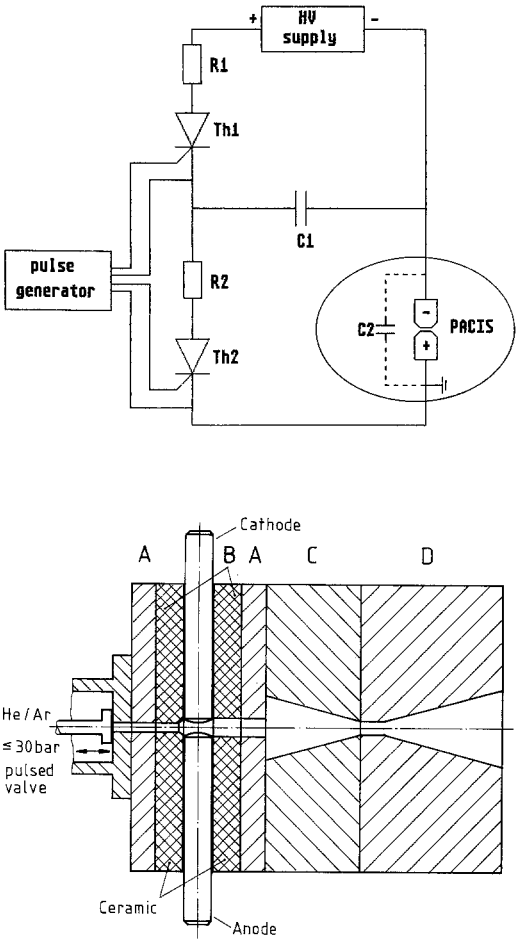


**Fig. 1. a** Power supply using thyristors to initiate the discharge in the PACIS. The capacity $C_1$ is charged and discharged by delayed triggering of two thyristors. A voltage up to 1300 V and capacities up to 23.5 µF are used. In some cases an additional capacity ($C_2 \simeq 1/3\ C_1$) is added parallel to the electrodes. **b** Design of the PACIS. The discharge is fired between two isolated electrodes in the presence of a carrier gas. A: aluminium plates, B: insulator block (BN or Macor), C: "mixing chamber", D: extender with conical nozzle

source chamber pumps are throttled in order to allow an increase of the background pressure to 1 mbar. Under these conditions a Mach disk is expected between source and skimmer at a distance $x_M$ downstream the source [14]:

$$x_M = 0.67 \cdot d \cdot (p_0/p_1)^{1/2} \quad (1)$$

with $d$ the orifice diameter, $p_0$ the stagnation pressure, i.e., the pressure just upstream the orifice, and $p_1$ the background pressure.

Under the condition of large discharge power and without any extender or mixing chamber, a significant portion of the plasma mixture remains electronically excited. Thus intense light is emitted from the expanding jet, clearly showing a barrel shock and a Mach disk. Apart from the possibility of studying expansion dynamics with this method, we can use the position of the Mach disk and (1) to estimate the pressure at the exit of the source. In good agreement with the above calculation a value of 1.4 bar is obtained.

A more detailed discussion of the source performance parameters and a comparison of the two power supplies will be given elsewhere [15].

## III. Results and discussion

Some results will be given to demonstrate the properties of the PACIS. First, the intensity of the source is estimated. Next, the build-up of a resolved mass spectrum from an unpulsed bunch will be shown to open a way to determine cluster beam velocities. Finally, some TOF mass spectra will be presented. If not stated otherwise, the following results have been obtained with the thyristor-driven source.

A quartz crystal microbalance is used to measure the overall intensity of emitted metal. The deposition rates clearly depend on the distance extender-target as well as on source conditions and electrode material. With a configuration of the PACIS as in Fig. 1b and at 5 cm distance, rates of 0.1 Å/shot and 1.8 Å/shot are obtained for Ag and Pb. Shot to shot variations were about 100%. Using deflection plates while depositing, e.g., Ag, the ion to neutral ratio can roughly be estimated to about 13 (+/−5)%.

The integral *ion* current is measured on and behind the (8 mm diam) skimmer, at 10 cm and 40 cm downstream the source nozzle, respectively. In order to increase the collection efficiency, a potential of a few volts may be applied either to the skimmer or to the collecting grid. By this, up to $2 \cdot 10^{12}$ negative ions per shot are measured on the skimmer, and about $2 \cdot 10^{11}$ on the collecting grid behind the skimmer. After changing the collection voltage to negative values positive ions are detected. Their intensity is roughly a factor of three lower compared to the negative ions. From the time dependence of the ion current measured without collection voltage a significant influence of the ion charge state on the flight time is observed. Negative carbon clusters, e.g., arrive about 40 μs earlier at the skimmer than positive ones. This might be due to the ionization and attachment processes in the source, or to mass distributions which are different for positive and negative ions. A qualitatively similar effect has been seen for electron impact ionized caesium clusters from a pure vapor expansion [16]. From the shift of the peaks with rising potential we learn that the measured ion currents are not much influenced by free electrons in the jet. With very low He pressure, however, up to $5 \cdot 10^{12}$ electrons per shot are measured at the skimmer biased to 50 V.

The build-up of a resolved $Pb_n^-$ mass spectrum is demonstrated in Fig. 2. Without switching the acceleration voltage ("grid pulse") in the TOFMS, the cluster ion bunch entrained in the carrier gas passes the spectrometer. The detected spectrum is displayed in Fig. 2 by the dotted curve. It shows a spread of the cluster bunch to about 500 μs (after a drift length of 1.4 m). When applying a 100 μs long grid pulse (300–600 V), a part of the former unaccelerated bunch will form a mass-resolved spectrum at shorter flight times. The remaining unaccelerated part

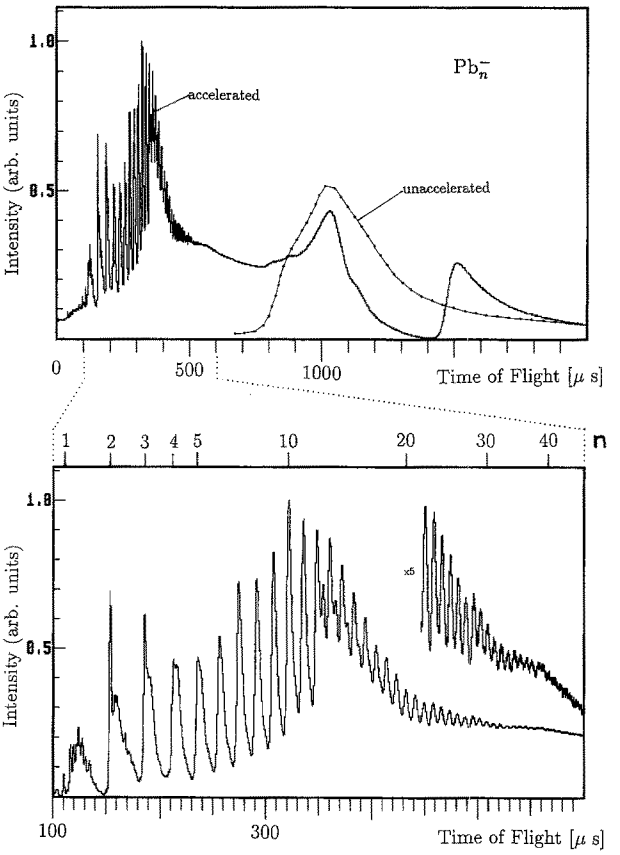


**Fig. 2.** TOF-spectra of $Pb_n^-$. The dotted line shows the detector signal without pulsing the TOF grids. When accelerating the clusters in the TOFMS, the former broad line displays a pronounced gap. In addition, the accelerated ions form a resolved mass spectrum, see left side and bottom picture

of the spectrum exhibits a broad dip. By varying the delay time between discharge and grid pulse, different portions of the cluster ion cloud can be analyzed. Regarding the involved velocity distributions (see below), the main parts of the cluster bunches leaving the PACIS have a length of at least 150 μs, followed by even longer tails. It should be noted that a broad ion bunch like this does not necessarily mean a missing narrowing effect of the velocity distribution in the free jet expansion; the ions are generated over more than 10 μs and are thermalized in the mixing chamber (Fig. 1b), thus diluted over a large time range.

How can we get information about the velocity of the ion pulse from such investigations? The part of the unpulsed spectrum just in front of the dip (see Fig. 2) represents those cluster ions which have already left the acceleration region at the time of the grid pulse. Therefore, we can calculate the average velocity of the cluster ions at the beginning and at the end of the grid pulse. Evaluation of Fig. 2, e.g., yields velocities between 1360 and 1100 m/s, respectively. These are the slowest clusters we have measured so far, but such velocity slip effects are already known in the literature [17, 18]. For a more fully relaxed expansion and thus reduced velocity slippage, Ar or Ne carrier gas would certainly be the better choice. Nevertheless, with changed source conditions Pb-velocities of

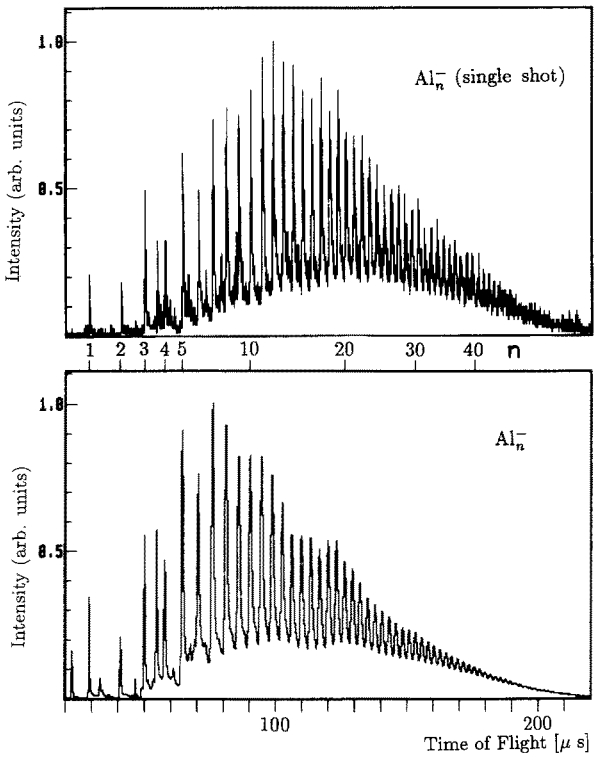


**Fig. 3 a, b.** TOF-spectra of $Al_n^-$. Here $LN_2$-cooling has been supplied, yielding an increased intensity and better reproducibility. **a** Typical single shot spectrum. The envelope and structure may change from shot to shot. **b** Averaged spectrum of about 100 shots under the same source conditions

about 1700 m/s are received for the low-mass clusters. In the case of large $Al_n^-$ the measured velocity ranges from 1600–1860 m/s.

Figure 3 shows a typical single shot mass spectrum, with the envelope of the individual shots varying to a certain extent. Eventually some lines with enhanced intensity appear, but generally the averaged spectra show nearly smooth overall envelopes. This holds true for all metals analysed so far, e.g., Ag and W [1, 2], and for both positive and negative ions. Carbon clusters are an exception under all source conditions, mostly yielding the typical "magic numbers" 11, 15, 19, 23 . . .

In summarizing, the PACIS developed only a year ago has now proved to be a reliable tool to produce jet-cooled clusters of conductive solid materials. The intensities and size distributions are comparable to those obtained with a laser vaporization source. Furthermore, no additional ionizing agent is needed to receive high ion currents of both positively and negatively charged clusters.

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