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Ion mass and energy selective hyperthermal ion-beam assisted deposition setup

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For the synthesis of high-quality thin films, ion-beam assisted deposition (IBAD) is a frequently used technique providing precise control over several substantial film properties. IBAD typically relies on the use of a broad-beam ion source. Such ion sources suffer from the limitation that they deliver a blend of ions with different ion masses, each of them possessing a certain distribution of kinetic energy. In this paper, a compact experimental setup is presented that enables the separate control of ion mass and ion kinetic energy in the region of hyperthermal energies (few 1 eV – few 100 eV). This ion energy region is of increasing interest not only for ion-assisted film growth but also for the wide field of preparative mass spectrometry. The setup consists of a constricted glow-discharge plasma beam source and a tailor-made, compact quadrupole system equipped with entry and exit ion optics. It is demonstrated that the separation of monoatomic and polyatomic nitrogen ions (N^+ and N_2^+) is accomplished. For both ion species, the kinetic energy is shown to be selectable in the region of hyperthermal energies. At the sample position, ion current densities are found to be in the order of $1 \mu A/cm^2$ and the full width at half maximum of the ion beam profile is in the order of 10 mm. Thus, the requirements for homogeneous deposition processes in sufficiently short periods of time are fulfilled. Finally, employing the described setup, for the first time in practice epitaxial GaN films were deposited. This opens up the opportunity to fundamentally study the influence of the simultaneous irradiation with hyperthermal ions on the thin film growth in IBAD processes and to increase the flexibility of the technique. *Published by AIP Publishing.* [<http://dx.doi.org/10.1063/1.4985547>]

I. INTRODUCTION

Ion-beam assisted deposition (IBAD) is a hybrid technique based on physical vapor deposition (PVD) processes, where the growing film is subjected to a continuous or periodic irradiation with a flux of energetic particles. IBAD on the one hand offers the opportunity to investigate fundamental processes involved in ion-assisted thin film growth and on the other hand provides manifold possibilities to intentionally modify the properties of the thin films fabricated (for details, see reviews in Refs. 1–4). By taking advantage of the interaction of the simultaneously impinging energetic ions with the near surface region of the growing film, the film growth processes as well as the resulting film properties can be controlled. In contrast to other prominent PVD techniques, e.g., magnetron sputtering, IBAD techniques are mainly defined by spatially separated material fluxes, which are directed towards the sample. Moreover, these material fluxes, i.e., vapor flux and ion flux (usually given in combination as the so-called ion-to-atom ratio), can be accurately and independently adjusted. The ion flux is usually generated in the form of a broad ion beam produced by an ion-beam source. Typical ion energies used in IBAD are in the keV range. Apart from the preparation of pure single-element films using inert noble gas ion bombardment, IBAD is well suited for the deposition of compound thin films, such as nitride or oxide films, by using reactive ion species.

This is another consequence of the independently adjustable material fluxes.

Despite its widespread use, IBAD with conventional broad-beam ion sources (Kaufman type, radio frequency excited, electron cyclotron resonance type, etc.)^{1–5} is characterized by several significant limitations originating from the properties of the respective ion-beam source as follows:

- (i) The ion beams from broad-beam sources for reactive gas species like nitrogen, typically generated by nitrogen plasma based ion-beam sources, contain a blend of both molecular (polyatomic) nitrogen ions and atomic (monoatomic) nitrogen ions, N_2^+ and N^+ , respectively.^{6,7} Their ratio can only be adjusted in a narrow range without altering other beam parameters; the generation of an ion beam consisting of either atomic or molecular ions is unfeasible with such typical broad-beam ion sources.
- (ii) Each of the molecular and atomic ion species in the beam possesses an independent energy distribution, which cannot be tuned individually. In the ion beam, which hits the film surface, all components overlap, leading to a broad energy distribution with several maxima.^{6,7} This impedes the precise control over the film growth processes and can lead to the deterioration of the resulting film properties, for

instance, due to ion-irradiation induced defect formation.

- (iii) Another issue is obtaining ions with low kinetic energy at sufficiently high ion beam current, particularly in the case of hyperthermal ion energies, ranging from a few eV to a few 100 eV.^{8–12} This leads to insurmountable difficulties in assessing the dissemination of the ion energy to the growing film surface in a well-defined, precise, and reproducible way. And this applies especially to the case of ion-beam assisted molecular beam epitaxy (IBA-MBE), where the use of hyperthermal ions with energies of only a few 10 eV is required.^{13,14}

In this publication, a compact setup for ion mass and ion energy selective IBAD is presented, which allows creating a nitrogen ion beam of variable hyperthermal ion energy, with independently selectable ion mass, at ion beam currents which are sufficiently high for the purpose of thin film deposition. Integrated into an IBA-MBE system (see Ref. 15 for details), the setup was realized by coupling a constricted glow-discharge plasma beam source¹⁶ with a custom built radio-frequency (RF) quadrupole mass filter, equipped with entry and exit ion optics, ion-beam deflection, and ion-beam current monitoring. In contrast to other reported setups for the production of mass-selected hyperthermal ion beams applied, e.g., for ion beam deposition and ion-solid collision experiments,^{8,10} the setup presented here is comparably compact, as it is not a beam-line setup. In addition, as the presented setup is meant for application in thin film deposition experiments, as opposed to typical beam-line setups, it is optimized for high ion current densities at a comparably large sample area. In Secs. II–V, the ability of the presented ion mass and energy selective setup to provide intense hyperthermal, mass filtered molecular or atomic ion beams extracted from reactive gases is demonstrated. The behavior of the ion beam in terms of intensity, beam spread, space charge, and purity is described. Finally, the feasibility for hyperthermal ion-beam assisted growth with mass and energy selected ions is demonstrated by fabricating high-quality thin films of gallium nitride (GaN) deposited epitaxially on single-crystalline substrates at elevated temperatures.

II. MASS FILTER SETUP

Ion mass filtering can be achieved by applying various principles. Here, the conditions to be taken into account are (i) mass resolution sufficiently high to separate atomic and molecular nitrogen ions, (ii) low ion energy in the range of hyperthermal energies, (iii) ion beam current high enough for ion-assisted deposition processes, and (iv) broad ion beam with a beam diameter in the order of 10 mm. Due to the reliance on small apertures, the use of mass filters based on magnetic fields, e.g., Wien filters or magnetic sector field mass filters, typically leads to significant losses of ion beam current, especially in the case of low ion energies.^{10,17} The same argument holds true for electrostatic mass filters. Quadrupole mass filters, however, which rely purely on electric fields, provide a compact and efficient setup for the mass separation of hyperthermal ions at comparably high ion beam currents.

Therefore, a quadrupole mass filter setup as depicted in Fig. 1 is employed in the context of this work. It consists of a source region with the plasma source and extraction electrodes, the RF-quadrupole mass filter, and finally DC ion optics. The constricted DC glow-discharge (hollow-anode) plasma beam source,¹⁶ which is a filamentless type of plasma source, is positioned approximately 2 cm in front of the entry electrodes of the setup, a series of 5 ring electrodes (labeled as 1-5 in Fig. 1). The first electrode (1) is equipped with a stainless steel grid that possesses a transparency of 70%. The purpose of this grid with an applied DC voltage U_{grid} is (i) to shield the entry region from the plasma and (ii) to support the ion extraction from the plasma. Together with an additional lens (6), the following stack of entry electrodes (2-5) couples the extracted ions into the quadrupole and focuses the generated ion beam close to the quadrupole axis. The following set of quadrupole rods is divided into three segments (7-9), each being characterized by a field radius $r_0 = 8.75$ mm and a rod radius $r = 10$ mm. The ratio of the two radii was chosen within the interval for optimum mass (m/z) resolution of $1.14 \leq \frac{r}{r_0} \leq 1.16$.¹⁸ Moreover, the used ratio of the radii is close to the optimum value of 1.145 for which the parasitic 12-pole term of the electric field of four circular electrodes vanishes. In all three segments, an RF voltage of constant frequency (3 MHz) but variable amplitude of up to 1.2 kV is applied to the rods at opposite polarity for neighboring rods. The operation frequency was chosen as a compromise between the electric losses and the depth of the effective potential (see Sec. VI for more details). Generally, for a given ion mass, a higher operation frequency requires a higher operation voltage. This provides a deeper effective potential, thus better ion confinement. Since the power dissipation in the system is proportional to the operation frequency and the square of the operation voltage, it increases strongly with the chosen frequency.¹⁹ The selected operation parameters (3 MHz and 1.2 kV) were identified as maximum achievable values for a long-term operation of the quadrupole system.

In addition to the RF potential, DC potentials are applied to each segment, separately. For the first (7) and the third (9)

FIG. 1. Schematics and a photograph of the quadrupole mass filter setup (from left to right): ion extraction grid, set of entry electrodes, three separate quadrupole segments, set of ion optics electrodes, and beam current monitoring grid.

quadrupole segments, a DC potential is applied to all four rods and is determined by the neighboring entry (6) and exit (10) electrodes, respectively. Consequently, these DC potentials are called U_{entry} and U_{exit} . Being operated solely in the RF-only mode, the first and third quadrupole segments (7 and 9) function only as ion guides. In addition to the DC potential U_{axis} of the center quadrupole segment (8), a differential DC voltage (opposite polarity between neighboring rods) can be applied. Consequently, this quadrupole segment can operate as a mass (m/z) filter. This implementation of separate quadrupole segments with their respective function was suggested by Dawson to decrease the loss of ions when coupling into and out of the mass filtering quadrupole by reducing the influence of fringing fields.²⁰

After passing the quadrupole segments, the ions enter a set of ion optics electrodes (10–13) that allow focusing as well as deflecting the ion beam. Finally, the ion beam passes a high transparency (96%) stainless steel grid (14), which is used to monitor the ion current leaving the setup and to shield the drift region between this grid and the sample location from electric fields that originate from the electrodes within the setup. The total length of the quadrupole system, measured from the ion extraction grid (1) to the ion current monitoring grid (14), is approximately 55 cm, where the center quadrupole segment is the longest component of the system with a length of 34.6 cm.

The constricted glow-discharge plasma source delivers a streaming nitrogen plasma containing hyperthermal atomic (N^+) and molecular ions (N_2^+) with kinetic energies less than 25 eV.⁶ The source is typically operated with a nitrogen gas flow of 2.5 standard cubic centimeters per minute (scm), which results in a pressure of 4×10^{-2} Pa close to the first segment of the quadrupole and around 3×10^{-3} Pa in the drift region between the quadrupole setup and sample location.

III. ION MASS SELECTION

The general working principle of linear RF quadrupole mass filters has been investigated extensively (see, e.g., the reviews in Refs. 21 and 22). An RF potential with amplitude U_{RF} and a differential DC potential U_{DC} are applied to the quadrupole rods, such that the voltage between neighboring rods $V(t)$ is given by

$$V(t) = 2U(t) = 2[U_{\text{DC}} + U_{\text{RF}} \sin(\omega t)], \quad (1)$$

with the radio frequency being $f = \frac{\omega}{2\pi} = 3$ MHz in the present setup. Ideal quadrupole rods are hyperbolic in profile, which is approximated by a semicircular profile in the presented setup. Assuming a hyperbolic profile, the potential distribution $\phi(x, y)$ on a cross section of the quadrupole can be expressed as follows:

$$\phi(x, y) = U(t) \frac{x^2 - y^2}{r_0^2}, \quad (2)$$

where r_0 is the quadrupole field radius. The resulting equations of motion for ions in this field can be rewritten as Mathieu's

differential equation using the following parameter definitions:

$$a = \frac{8zeU_{\text{DC}}}{m\omega^2 r_0^2}, \quad (3)$$

$$q = \frac{4zeU_{\text{RF}}}{m\omega^2 r_0^2}, \quad (4)$$

where ze is the charge of the respective ions and m is their mass. Depending on the parameters a and q , the ion trajectories are either stable, such that entering ions are transmitted through the quadrupole, or unstable, meaning that the ions gain energy from the electric field, thus increase their oscillation amplitude, and leave the electrode system or collide with an electrode. This can be summarized in the stability diagram by plotting parameter a versus q , which displays a characteristic, triangular-like shaped region of stable trajectories for small values (<1) of a and q . According to Eqs. (3) and (4), such a stability diagram can also be given as a function of U_{RF} and U_{DC} , for the given mass, charge, frequency, and quadrupole geometry, where the stable regions are shifted according to the mass to charge ratio of the ions.

Figure 2 displays a corresponding measurement of the ion current, measured with a Faraday cup (diameter of orifice: 5 mm) at the sample position, in dependence of the DC potential U_{DC} and the RF amplitude U_{RF} . Two main contributions are visible in this diagram, denoted as N^+ and N_2^+ , referred to as atomic and molecular nitrogen ions. Additionally, the stability regions of N_2^{2+} and the isotope $^{15}N^+$ can be detected. As the plasma source is operated with nitrogen gas that contains impurities of argon, a stability region for Ar^{2+} can be discerned too.

Furthermore, corresponding to the analytical stability diagram, the border lines of regions, where the trajectories of these ions would be stable, are indicated by dashed white lines, as

FIG. 2. Ion beam current, measured with a Faraday cup at the sample position, plotted in dependence of the DC potential U_{DC} and the amplitude of the RF potential U_{RF} applied to the center quadrupole rods (8). In this measured stability diagram, the triangularly shaped stability regions of the most prominent ion species are denoted. The dashed white lines correspond to the calculated border lines of the regions of stable ion trajectories. The dashed black line indicates a typical mass scan line, on which specific ion species are selected. The black circles indicate typical chosen operating conditions for the transmission of N^+ and N_2^+ , respectively.

calculated from the polynomial solution resulting from Mathieu's differential equation.²³ The calculated stability regions are in good agreement with the measured data. Hence, it is demonstrated that the presented setup is capable of separating different ion species, particularly atomic and molecular nitrogen ions, and therefore can function as a mass filter. Typically, quadrupole mass filters are operated along the so-called mass scan lines, where U_{DC} and U_{RF} are varied while keeping their ratio constant. The value of this ratio determines the mass resolution ($m/\Delta m$) of the mass filter, where the optimal mass resolution is achieved by passing through the tips of the stability regions. Here, the mass resolution only needs to be high enough to achieve the separation of atomic and molecular ions. Since mass resolution and transmission counteract each other,²⁴ the quadrupole is operated under the chosen condition $U_{DC} = 0.1U_{RF}$ in order to allow for reasonably high ion beam currents while separating molecular and atomic nitrogen ions.

IV. ION ENERGY SELECTION

In the following, the distribution of the kinetic energy of the ions transmitted through the quadrupole setup is examined, as measured using a Faraday cup with a retarding field grid located at the sample position. It was found that both the magnitudes of U_{entry} and U_{axis} define the kinetic energy of atomic and molecular ions as shown in Fig. 3. It is exemplarily shown that increasing U_{entry} , while keeping the other potentials and conditions constant, accordingly increases the average kinetic energy of atomic ions. Additionally, for molecular ions, the kinetic energy is increased by increasing U_{axis} . The distributions of kinetic energy are reproduced by Gaussian functions. The full width at half maximum (FWHM) for the main contribution to the kinetic energy distribution of atomic ions and molecular ions is approximately 20 eV and 10 eV, respectively. Especially for molecular ions, there are deviations from the Gaussian shape on the higher-energy tail of the peak, showing that molecular ions with higher kinetic energies also contribute. The widths of the distributions however are small enough to allow for investigating the basic influence of the ion kinetic energy on film growth processes.

The explanation for the observed kinetic ion energy distributions and for the identified dependencies lies in the combination of the plasma source and the quadrupole system presented here. Initially, the ions are, due to a pressure gradient, gas-dynamically accelerated in the nozzle of the plasma source.²⁵ The resulting kinetic energy of the ions in this streaming plasma is smaller than 25 eV.⁶ The related ion energy distributions of ions from the plasma source entering the quadrupole system are given and discussed elsewhere.⁶ In the region of the entry electrodes (1-6) and also of the first two quadrupole segments (7 and 8), the ions are extracted from the plasma as well as from the high pressure region, which means that the influence of the plasma space charge as well as of the collisions with neutral gas molecules decreases to the point where they do not play a role. Since atomic and molecular nitrogen ions have a different ion mass and also a different cross section for collisions due to their different sizes, their

FIG. 3. Distributions of kinetic energy for (a) atomic ions varying U_{entry} and (b) molecular ions varying U_{axis} , as determined at the sample position by using a Faraday cup equipped with a retarding field grid. For the measurement plotted in black in (b), the potentials of the entry electrodes were also changed, in particular, the potential of the ion extraction grid (1).

behavior in that particular section of the quadrupole differs significantly. Therefore, the extraction from the plasma and from the region dominated by collisions between the ions and the background gas can take place in different positions depending on ion species and the operation conditions of the plasma source and the quadrupole system. In principle, the ion energy selection works in the following way: The ions enter the region of a certain electrode with a positive potential. The potential is screened due to the plasma or collision with the background gas which dominates the behavior of the ion trajectories, such that there is no effective potential barrier to decelerate the ions. Within this electrode, the ions are extracted. When the ions leave the region of the electrode, a potential drop accelerates them. Hence, their kinetic energy is defined depending on the potential at the extraction location. In conclusion, the kinetic energy of the mass-selected atomic and molecular ions [see also Fig. 4(b) in Sec. V] reaching the sample position can be adjusted and controlled within the typical range of hyperthermal ion energies (see Sec. I) with sufficient accuracy.

FIG. 4. Characteristics of the ion beam after leaving the quadrupole system: (a) Ion current measured on a Faraday cup with an area of $1\text{ cm} \times 1\text{ cm}$ (red) and on a disc with a diameter of 5 cm (black) divided by the total ion beam current measured by the beam monitoring grid (14), plotted in dependence of the distance from the quadrupole exit. (b) Achievable ion current densities plotted in dependence of the average ion kinetic energy for atomic ions (black) and molecular ions (red) at a distance between the Faraday cup from the quadrupole exit of 7.5 cm , corresponding to the sample position. (c) Shape of the ion beam at a distance of 7.5 cm from the quadrupole exit, measured by moving a Faraday cup across the ion beam in two perpendicular directions (average kinetic energy of atomic ions of 40 eV). (d) The shape of the ion beam for identical average kinetic energy as simulated with SIMION. The inset shows 10 000 ion trajectories for optimized ion optics electrode potentials as indicated next to the respective electrode.

V. ION BEAM CHARACTERISTICS

The characteristics of the ion beam leaving the quadrupole system are of importance with regard to the homogeneity and duration of the film deposition process as well as the quality of the resulting thin films. Figure 4 sums up the relevant properties of the ion beam: achievable ion beam currents and ion current densities, ion beam profile, and divergence of the ion beam. According to the measurement [red points in Fig. 4(a)], the ion current measured on a Faraday cup with an area of $10\text{ mm} \times 10\text{ mm}$ (equal to sample size) decreases with increasing distance from the quadrupole exit. This is an indication for the presence of ion beam divergence. Using an ion current measurement device with a larger detection area in the form of a disc with a diameter of 5 cm , it is expected that the measured ion current of a divergent ion beam remains constant up to a certain distance from the quadrupole exit. For distances exceeding this specific distance, the ion beam spread gets larger than the dimension of the measuring device and the measured ion current starts to decrease. This behavior can be indeed observed in Fig. 4(a) (black measurement points). For

a distance from the exit of the quadrupole of 7.5 cm , approximately 90% of the maximum available ion beam current is collected by this disc. Assuming a Gaussian ion beam shape, the FWHM of the ion beam profile can be approximated to be 3.5 cm , accordingly. Due to the observed beam divergence, it is beneficial to decrease the distance between the exit of the quadrupole and the sample position as much as possible, so that both the ion current densities and the resulting film deposition rates are sufficiently high. The chosen operation distance of 7.5 cm is a compromise of this requirement and the necessity that the Ga atom flux from the effusion cell can reach the sample position without being shadowed by the quadrupole exit.

Figure 4(b) illustrates achievable ion current densities at this chosen distance of 7.5 cm from the quadrupole exit to the sample position. The corresponding total ion beam currents I_{atomic} and $I_{\text{molecular}}$ measured at the grid for ion current monitoring for the data points shown lie in the ranges of $0.2\text{ }\mu\text{A} < I_{\text{atomic}} < 3.9\text{ }\mu\text{A}$ and $0.1\text{ }\mu\text{A} < I_{\text{molecular}} < 0.9\text{ }\mu\text{A}$, depending on the kinetic energy of the respective ions. Clearly, the ion current densities increase with increasing ion kinetic energy. It

is known from computer simulations that ions are lost depending on the number of RF cycles and that for this reason, the transmission of a quadrupole increases with increasing kinetic energy of the ions.²⁶ Further, at higher kinetic energies, the space charge is lower at the same ion current. This may also increase the transmission of the system.

According to the so-called Brice model,^{13,27} the optimum kinetic energy of atomic nitrogen ions assisting the deposition of GaN should be approximately in the range $20 \text{ eV} \leq E_{\text{opt}} \leq 50 \text{ eV}$ in order to obtain a high crystalline quality of the resulting films. If this requirement is fulfilled, the maximal achievable ion current density is $j \sim 0.5 \frac{\mu\text{A}}{\text{cm}^2}$ [as extracted from Fig. 4(b)]. In the case that all impinging ions remain at the sample surface and contribute to the growth of the nitride film, deposition of a film with a thickness of 10 nm is achievable within a few hours. For molecular nitrogen ions, however, it can be assumed that an additional amount of energy is necessary to dissociate the molecular ion. Consequently, the remaining kinetic energy is divided between the two resulting atoms which would each have just below half the energy. The kinetic energy of molecular nitrogen ions therefore can potentially be increased accordingly, reducing the time needed for film growth of identical thickness.

In order to ensure the deposition of films with high homogeneity, the FWHM of the broad ion beam has to match the size of the sample. Figure 4(c) shows the ion beam shape measured by moving a Faraday cup with a high lateral resolution (diameter of orifice $d = 1 \text{ mm}$) through the ion beam in two perpendicular, cross-sectional directions for atomic ions with an average kinetic energy of 40 eV. On the shown length scale, the beam shape can be approximated by a Gaussian function. The FWHM for the two lateral directions are 12 mm and 11 mm, respectively. Evidently, the ion beam cross section is rather circular and thus does not exhibit significant ellipticity. The ellipticity inherently originating from the elliptical shape of the effective potential in the mass-filtering quadrupole segment (see below) obviously vanishes behind this segment. Furthermore, it can be concluded that homogeneous film deposition is achievable on length scales in the order of several mm. Figure 4(d) shows the result of an ion trajectory simulation using the SIMION software.²⁸

Ion trajectories in the three quadrupole segments were simulated in order to estimate the angular and spatial distribution of ions leaving the last quadrupole segment and entering the ion optics. Assuming these as the starting conditions for a simulation of the ion trajectories in the ion optics, the potentials applied to the ion optics electrodes were optimized such that the number of ions arriving on the sample with less than 2.5 mm distance from the ion beam axis was maximized. For these optimized potentials, the spatial distribution at a distance from the quadrupole exit corresponding to the sample position is shown for ions with an average kinetic energy of 40 eV. The indicated red and black fit curves are each a sum of two Gaussian functions with FWHM values of 11 mm and 12 mm for the two narrow functions and 37 mm and 38 mm for the broader ones. These simulated data are in good agreement with the measured data of the beam shape on small length scales [see Fig. 4(c)] and the beam divergence approximated from Fig. 4(a) on larger length scales.

High ion currents, which are required for a preparative application of a quadrupole mass filter, suggest that space charge repulsion can be the cause of beam broadening and a limiting factor for the achievable ion current.²⁹ However, the simulation reproduces the measured beam profile and the beam divergence well without taking into account space charge effects. Therefore, a reason for the divergence is assumed to be the broad angular distribution of the trajectories of the ions, which are coupled out of the RF potential.

VI. ESTIMATIONS REGARDING SPACE CHARGE EFFECTS

In the present setup, space charge limitations are considered for two regimes: the trapped ion beam within the quadrupole segments and the free ion beam between the exit lens and the substrate.

The ion motion in the quadrupole can be described by the solution of Mathieu's differential equation that is in general an infinite series of harmonic oscillations. The first one with the lowest frequency is called secular motion and has the frequency $\Omega = \frac{1}{2}\beta\omega$, where β is a number depending on the parameters a and q .³⁰ An approximate formula³¹ for β is given by

$$\beta \cong \sqrt{a - \frac{(a-1)q^2}{2(a-1)^2 - q^2} - \frac{(5a+7)q^4}{32(a-1)^3(a-4)} - \frac{(9a^2+58a+29)q^6}{64(a-1)^5(a-4)(a-9)} + \dots} \quad (5)$$

The ion motion is stable for values of β between 0 and 1; these values determine the borders in the stability diagram. Comparing the harmonic oscillations with the secular frequency to a linear mechanic oscillator, an equivalent potential, the so-called effective potential, can be calculated as

$$\phi_{\text{eff}} = \frac{\beta^2 m \omega^2 r^2}{8ze}. \quad (6)$$

The coordinate r is the radial distance from the quadrupole axis.

The entry and exit segments of the quadrupole are operated in the RF-only mode, i.e., with $a = 0$. Under the assumption of an adiabatic ion motion, i.e., low values of parameter q , the following approximation for β can be used:

$$\beta = \frac{1}{\sqrt{2}} |q| = \frac{2\sqrt{2}zeU_{\text{RF}}}{m\omega^2 r_0^2}, \quad (7)$$

implying the effective potential

$$\phi_{\text{eff}} = \frac{zeU_{\text{RF}}^2}{m\omega^2 r_0^2} \left(\frac{r}{r_0} \right)^2. \quad (8)$$

The middle quadrupole segment is operated as a mass filter, i.e., with a DC voltage ($a \neq 0$). The polarity of the DC voltage is opposite for both directions resulting in opposite signs of parameter a . Compared with the RF-only mode, the values of β are lowered in one direction and increased in the other one; they reach 0 or 1, respectively, on the boundary of the stability region. Thus, the effective potential is not symmetrical but deeper in one direction and flatter in the other one.

The influence of the effective potential on the transmission can be derived from the measurements in Fig. 2. The left-side boundary of the stability region corresponds to $\beta = 0$, i.e., $\phi_{\text{eff}} = 0$; thus, at this boundary, the ions are not confined at all. The higher the distance from the boundary, the deeper is the effective potential and the stronger is the ion confinement. Since the measured output current reaches the maximum value already in the proximity of the above-mentioned boundary, the system apparently does not lose ions due to the insufficient depth of the effective potential in the mass-filtering segment.

For the other two segments operated in the RF-only mode, the effective potential [Eq. (8)] is estimated to be $80 \text{ eV} < \phi_{\text{eff}} < 125 \text{ eV}$ for atomic nitrogen ions assuming $U_{\text{RF}} = 700 \text{ V}$. The radial distance is chosen to be in the range of $0.8 < \frac{r}{r_0} < 1$ because of deviations from the ideal electric field near the quadrupole rods. These values for the effective potential can be compared to the space charge potential of the beam ϕ_{beam} , which can be estimated as

$$\phi_{\text{beam}} = \frac{I}{4\pi\epsilon_0} \sqrt{\frac{m}{2E_{\text{kin}}}}, \quad (9)$$

where E_{kin} is the kinetic energy of the ions. This expression is derived when integrating the space charge fields of a uniform and circular beam with the radius of the beam set equal to the field radius.³²

For the given setup assuming atomic nitrogen ions with a kinetic energy of 40 eV and a total ion current of $100 \mu\text{A}$, the space charge potential is $\phi_{\text{beam}} \cong 40 \text{ eV}$. Therefore currents of up to $100 \mu\text{A}$ can be transmitted by the quadrupole segments in the RF-only mode, since the space charge is compensated by the quadrupole's effective potential.

To estimate the beam spread and the relevance of space charge for the ion beam after leaving the ion optics, the perveance

$$K = \frac{q\sqrt{m}}{4\sqrt{2}\pi\epsilon_0} \frac{I}{E_{\text{kin}}^{\frac{3}{2}}} \quad (10)$$

is defined.³² The value of the perveance describes the influence of the space charge effects on the motion of the charged particles. Assuming a uniform ion current density in the ion beam and cylindrical symmetry with the direction of motion as the z -coordinate, the equation of transverse motion for an ion at the beam boundary (distance r_{env} from the beam axis) can be written using the perveance³²

$$\frac{d^2 r_{\text{env}}(z)}{dz^2} = \frac{K}{r_{\text{env}}(z)}. \quad (11)$$

Solving this equation numerically with $\frac{dr_{\text{env}}}{dz}(0) = 0$ for atomic ions with $E_{\text{kin}} = 50 \text{ eV}$ yields 1.24, 2.97, and 12.44 for the ratio

$\frac{r_{\text{env}}(7.5 \text{ cm})}{r_{\text{env}}(0)}$, assuming total beam currents of $1 \mu\text{A}$, $10 \mu\text{A}$, and $100 \mu\text{A}$, respectively. Accordingly, a significant beam spread due to space charge effects for a typical ion beam used here can be expected for ion currents in the order of $10 \mu\text{A}$ or higher. This confirms the observation, where the beam spread (for a typical beam current $I \sim 1 \mu\text{A}$) is attributed to the broad angular distribution of the trajectories of ions being coupled out of the RF potential.

Summarizing the previously described considerations for the design of the presented instrument, the relevance of space charge effects was evaluated for two regimes: the trapped ion beam within the quadrupole mass filter and the free ion beam in the region between the quadrupole exit and the sample position. To estimate the order of magnitude at which space charge limits the transmission of the quadrupole, the depth of the effective potential of the quadrupole segments in the RF-only mode is compared to the space charge potential of the beam. It is found that the transmission will not be affected by space charge, if ion currents are below $\sim 10 \mu\text{A}$. Moreover, from the ion current measurements shown above (Fig. 2), it can be deduced that in the mass-filtering quadrupole segment, ion currents of up to $\sim 4 \mu\text{A}$ are confined in the effective potential. The spread of the free ion beam is estimated for a distance of 7.5 cm, as defined by the given experimental setup, based on the beam's perveance. The calculations show that only a slight broadening can be expected for ion beam currents in the order of $1 \mu\text{A}$. For higher ion beam currents, the beam broadening starts to become significant.

The measured ion currents of up to $\sim 4 \mu\text{A}$ therefore suggest that the space charge does not play a major role, neither for losses in the quadrupole nor for the observed beam spread. Indeed the trajectory simulations show that coupling ions out of the RF potential can be sufficient to explain the observed beam spread. Based on these estimations, the achievable ion currents could be further increased by optimizing the coupling of the ion beam from the source into the quadrupole mass filter.

VII. SYNTHESIS OF EPITAXIAL GALLIUM NITRIDE

In this section, first results on the application of the presented setup for the mass selective hyperthermal ion-beam assisted deposition of epitaxial films are exemplarily shown. Depositions of GaN were carried out using either atomic or molecular nitrogen ions with the maximum of the kinetic ion energy distribution at 80 eV, ion current densities of $0.7 \mu\text{A}/\text{cm}^2$ and $1.2 \mu\text{A}/\text{cm}^2$, respectively, and at a 6H-SiC(0001) substrate temperature of 700°C . Reflection high-energy electron diffraction (RHEED) measurements were performed before and during the deposition (*in situ*). The RHEED patterns in Figs. 5(a)–5(c) represent the surface structure of the pristine 6H-SiC(0001) substrate, of the GaN film deposited with impingement of atomic nitrogen ions, and of the film deposited using molecular nitrogen ions, respectively. Comparison of the GaN film diffraction patterns with the calculated one in Fig. 5(d) reveals that (i) there is three-dimensional growth of epitaxial GaN, (ii) the deposited GaN possesses a pure cubic (zincblende) structure (z-GaN),

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