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By S. Kalbitzer & V. A. Zhukov

St. Petersburg Institute for Informatics and Automation, Russia

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Generation of Ion Nano-Beams with a Gas Field Ion Source

S. Kalbitzer ^a & V. A. Zhukov ^o

Abstract- With the advent of the super-tip gas field ion source very favourable conditions for generating ion nano-beams have been provided. The basic physical principles for this kind of source are described As a consequence of a very small source emittance of $\varepsilon \sim 10^{-24}$ m²sr ultra-bright rare gas ion beams of about 10¹⁶ A/m².sr can be produced. As a further consequence, very narrow probe sizes are obtainable, of d_p< 1 nm, with spatial resolutions far below the classical limits of light-optics and even below the novel stimulated-emission-depletion microscopy [STED (microscopy) = stimulated emission depletion (microscopy)] Th. A. Klar, S. W. Hell, Opt.Lett. 24 (1999) 954-956]. Various ion-nanobeam applications are envisaged.

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I. BASIC CONSIDERATIONS

his presentation summarizes our experimental work over a period of about 15 years carried out at the Max Planck Institut für Kernphysik at Heidelberg, Germany, started in 1984.

The first most important recognition in the above context had been that all conventional ion sources, in use for accelerators and most other physical equipments, are intrinsically useless for developping ion nanobeams because of their poor emittance figures. The liquid-metal-ion source, LMIS, has much better properties, but seems to be limited to probe sizes of the order of d_p ~ 100 nm.[1] Thus, the only remaining perspective for d_p ~ 1 nm has been to try to modify the existing Müller gas field ion microscope to an efficient ion source with very low emittance figures ϵ and high brightness B:

$$\varepsilon = \Delta A \Delta \Omega \sim d_0^2 \alpha_0^2, B = I/\varepsilon$$
 (1)

Where both the virtual source area ΔA of diameter d₀ and the emission solid angle $\Delta \Omega$ with bounding beam angles α_0 have to be many orders of magnitude smaller than those of standard ion sources. It is helpful, to discuss the enormous consequences of this feature. For simplicity, we consider the two limiting cases of purely chromatic and spherical aberration as shown by fig.1 in dependence on ε .

$$d_c = 2\eta^{1/2} C_c^{-1/2} \epsilon^{1/4}$$
 and $d_s = 2 C_s^{-1/4} \epsilon^{3/8}$. (2)

Thus, to achieve the dimensions of a nanometre beam we need a reduction of ε by many orders of magnitude: an atomic supertip.

In addition, there exists another quite difficult practical problem: previous pioneering attempts in this direction had not resulted in supertips stable over longtime operation periods.[2] In summary, the actual situation in 1984 did not look promising.

II. Realisation of the Super-Tip

A gas field ion source, GFIS, operates in a gaseous environment. Its main constituent is a sharply etched tungsten wire of some 100 nm radius. The demanding task is to fabricate an even smaller single protrusion on top of this tip as sketched in fig.2 to restrict field ion emission to a single truly nano-size emitter. The basic functional principles of the source involve adsorption of the respective gas species onto the tip surface and its subsequent surface diffusion to the tip apex, where ionisation takes place. Thus, in order to produce intense ion beams, a sufficient gas coverage of the tip surface constitutes a first prerequisit. Counteraction by thermal gas desorption from the tip surface has to be kept low. Effective measures are to either provide low enough temperatures and/or high electrical fields at the emitter tip enhancing both the incoming gas kinetic flux and the gas adherence to the tip surface. The Langmuir adsorption isotherm describes these processes :

$$\partial \theta / \partial t = j \cdot (1 - \theta) - v_{\rm H} \cdot \theta,$$
 (3)

$$v_{\rm H} = v_0 \exp(-H/kT), \ H = \frac{1}{2}\alpha F^2$$

where θ is the fractional occupation of the existing surface sites; j the incoming kinetic gas flux and $H = \frac{1}{2}\alpha F^2$ the energetic barrier to gas desorption, v_H the rate of desorption, $v_0 = 10^{13} \text{ s}^{-1}$ the lattice frequency. The fractional equilibrium occupation θ of surface sites (fig.3) is derived from the steady state condition $\partial\theta/\partial t = 0$:

 $\theta = 1/(1+\gamma), \gamma = v_H/j \text{ for any } \gamma \ge 0 \text{ and } \Phi = H/kT.$ (4)

Fig.4 displays the polarisation energies $E_p = \frac{1}{2} \alpha F^2$ for all rare gas species vs.electrical field strength. The emitted ionic current IC(kT, $E_{ps}, E_p)$ is plotted in fig.5 for an incident gas rate of $j = 10^{11}$ at/s, a physisorption energy $E_{ps} = 60$ meV and for several chosen values of induced polarisation energy E_p over a

Author α : Ion Beam Technology, Amselgasse 11, Heidelberg 69121, Germany. e-mail: skalbitzer@aol.com

Author σ : St. Petersburg Institute for Informatics and Automation, St Petersburg 199178, Russia.

range of thermal energies of kT = 10 - 25 meV. While a relatively low desorption barrier of H = 200 meV still leads to major desorption losses for room temperature operation, a barrier of H = 500 meV, however, nearly completely eliminates desorption. Low temperatures and/or high electrical fields support this process. Since conventional low temperature operation with liquid helium cooling is quite expensive, we check the other possibility:

An electric field of F = 20 V/nm, induces a polarisation energy of $E_p = \frac{1}{2}\alpha F^2 \sim 560$ meV for Xenon gas of polarisability $\alpha = 4.06 Å^3$, suffices to keep the Xe attached to the tip surface at room-temperature, but still mobile enough for diffusive transport to the tip apex, the diffusion sink.

The generation of an ion nano-beam, however, is mainly determined by the intrinsic source properties and only to a lesser extent by the optical transport system. All conventional ion sources have much too high emittance figures, so that the novel approach of developing a high quality source on the basis of field ionisation has been unavoidable. Virtual source size d₀ and angular spread α_0 of the emitted ion beam

determine the source emittance ϵ and also the properties of the resulting probe spot d_p:

$$\varepsilon = \Delta A \Delta \Omega \sim d_0^2 \alpha_0^2.$$
 (5)

with an observed emission angle of $1/2\alpha=8$ mrad and a calculated d₀ of 0.1 nm the result is $\epsilon\sim10^{-24}$ m²sr. [5] The corresponding brightness amounts to B=I/ $\epsilon\sim10^{16}$ A/m²sr.

Already with unfavorable values of $C_{\rm s} \sim 1$ m, $C_{\rm c} \sim 0.1 m$ and a relative energy spread of $\eta \sim 10$ ppm spot diameters in the low nm range are predicted. In fig.1 spot sizes $d_{\rm c}(\epsilon)$ and $d_{\rm s}(\epsilon)$ are plotted. A 4 o.m.reduction in ϵ is needed to yield 1 o.m reduction of $d_{\rm c}$ of the chromatic image spot size, what is impossible with conventional ion sources.

A second point of concern is the gas transport along the tip surface to the apex point.

Thus, for a tip radius of 600 nm the number of atomic hops n is derived as follows.

A diffusion length of 600 nm would lead to the result:

$$L=2(Dt)^{1/2}$$
 with $D=a^2v_0$ yields

$n = v_0 t = (L/2a)^2$ and with $a \sim 3$ Å: $n = 10^6$ diffusive hops.

During the diffusion time of t~100 ns the probability of desorption should be small. It is useful to introduce the variable $\Phi=\frac{1}{2}\alpha F^2/kT$, the ratio of the induced polarisation energy and thermal energy as a figure of merit. Fig.3 is an estimate employing desorption barriers in terms of $12 < \Phi \leq 20$, at 25 meV thermal energy. Small losses in θ occur for $\Phi > 20$, so that current levels of the order of the incoming atomic gas flux appear feasible as shown in fig. 5, permitting various applications in nanotechnology.

The probe current delivered to the emitter point is given by the GFIS*-"master equation":

$$J(z,\Phi) = j_0 \int \sigma \theta \eta \zeta dA(z) dz, \qquad (6)$$

where $j0\sigma(\Phi)$ denotes the field-enhanced gas kinetic gas flux to the tip surface, $\theta(\Phi)$ the steady state occupation of surface sites, $\eta(\Phi)$ the transport efficiency of surface atoms to the tip apex, ζ the ionisation efficiency of atoms delivered to the supertip apex, and dA(z)dz is the local surface element. Note that all factors in the integral of (6) are position dependent. Calculated emitter currents are displayed in fig.5.We see that a polarisation energy of $E_p \sim 500$ meV suffices to nearly completely deliver the incoming gas flux to the emitted ion beam. Now the remaining task is to develop an efficient ion beam optical system for both high beam transmission and nanometric resolution at still useful probe currents.

III. Applications of Ion Nanobeams

With energy as parameter, the range of ion interaction with the target material varies. Table 1 lists some basic properties of the available rare gas ion beams.

- a) Materials Analysis
- 1. Nano-scopic imaging. Surface topology is imaged by using secondary electron emission by ionic impact. He ions are to be preferred because of their high ionisation potential of about 25 eV leading to high electron emission figures. Spatial resolutions of better than 1 nm have been claimed for the Zeiss Orion device.[3].
- Nano-scopic surface chemical analysis by using secondary atom emission and time of flight mass spectrometry (TOF-SIMS) [4]. Here Xenon is the best choice because of its high sputter coefficients for all solids. Again, nm resolution is predicted.
- Excellent conditions for PIXE trace element analyses are provided with intense H₂⁺-nano-beams of MeV energy.
- b) Materials Modification
- Phase transitions in insulators, e.g. Si, from the crystalline to the amorphous state are readily induced with fluences of 10¹³ ions/cm² of the heavier rare gas species. Accompanying optical changes may be useful for optical memories of terabit storage capacity/cm² with permanent data life-time.

(5)

Reading of 10 nm bits, however, poses a severe problem.

 Nano-machining of material structures, e.g. pin holes, in solid surfaces by sputtering includes medical applications, e.g. cutting of biological material. Ne⁺ ion beams allow sputter action and simultaneous visual control of the crater by seondary electron detection.

3. Deposition of surface layers from injected gas environments near the target surface.

A recent review presents more details on these applied aspects.[5]

Z/A	$T_{c}(K)$	$\alpha(Å^3)$	X(eV)	v(cm/s)	$\Lambda(\text{fm})$	Y(W)	S	I(nA)
2/4	4.2	0.20	24.5	9x10 ⁵	143	~5	<0.2	2
10/20	27	0.39	21.5	4x10 ⁵	64	<4*	<0.8	2
18/40	87	1.62	15.7	2.8x10 ⁵	45	<3*	<3	2
36/84	120	2.47	13.9	2.0x10 ⁵	31	<2*	<2	2
54/13	165	4.06	12.1	1x10 ⁵	25	<2*	<10	2&
	Z/A 2/4 10/20 18/40 36/84 54/13	Z/A Tc(K) 2/4 4.2 10/20 27 18/40 87 36/84 120 54/13 165	$\begin{array}{c cccc} Z/A & T_c(K) & \alpha({\rm \AA}^3) \\ \hline 2/4 & 4.2 & 0.20 \\ \hline 10/20 & 27 & 0.39 \\ \hline 18/40 & 87 & 1.62 \\ \hline 36/84 & 120 & 2.47 \\ \hline 54/13 & 165 & 4.06 \\ \hline \end{array}$	Z/A $T_c(K)$ $\alpha(Å^3)$ $X(eV)$ 2/44.20.2024.510/20270.3921.518/40871.6215.736/841202.4713.954/131654.0612.1	$\begin{array}{c ccccc} Z/A & T_c(K) & \alpha({\rm \AA}^3) & X(eV) & v(cm/s) \\ \hline 2/4 & 4.2 & 0.20 & 24.5 & 9x10^5 \\ \hline 10/20 & 27 & 0.39 & 21.5 & 4x10^5 \\ \hline 18/40 & 87 & 1.62 & 15.7 & 2.8x10^5 \\ \hline 36/84 & 120 & 2.47 & 13.9 & 2.0x10^5 \\ \hline 54/13 & 165 & 4.06 & 12.1 & 1x10^5 \\ \end{array}$	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$

Table 1 : Some basic rare gas properties

⁸extrapolated value, at E ~1keV,* estimated by the relation ionisation energy/work function Y~X/W-1, W ~ 4.5 eV. T_{c} , gas condensation temperature; α , polarisability; X, ionisation potential, v ion velocity; Λ , de Broglie wavelength; *I*, emitter currents at $P \sim 0.1$ Pa; Y (electrons/ion), secondary electron yield; S (atoms/ion), sputtering yield; ion energy $E \sim 10$ keV.

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IV. FIGURE CAPTIONS



Figure 1 : Calculated image diameters dc(ϵ) and ds(ϵ) due to chromatic and spherical aberrations as a function of source emittance log(ϵ) = t. Note that an atomic size emitter of d₀ ~ 0.1 nm with a narrow emitter characteristic of $\alpha^{0} \sim 10$ mrad is needed for an emittance of $\epsilon \sim 10^{-24}$ m²sr and correlated image aberrations disks of d_i ~ 1nm. Parameters: $\eta \sim 10$ -5, C_c = 0.06 m, C_s = 1m



Figure 2 : Intended emission pattern of a regular 100 nm W-tip with a fictitious d₀ ~ 1 nm supertip on top and an emitter angel of $\frac{1}{2}\alpha_s \sim 10$ mrad



Figure 3 : Fractional occupation θ of surface sites for polarisation energies of $E_p = 200 - 500$ meV vs. tip temperature in units of meV. At the highest E_p the coverage remains very nearly complete: $\theta \approx 100\%$., over the entire temperature range up to 300 K



Figure 4 : Field induced polarisation energies E_p(F) for all rare gases vs. electrical field- strength F. The (dotted) demarcation line at 500 meV is to indicate the level of E_p necessary for a monolayer gas coverage of the enitter surface. Note that this level is most diffi¬cult to reach for He and Ne gas. Sequence from below upwards: He,Ne, Ar, Kr,.Xe



Figure 5 : Emitter current for varying source operation temperature and 5 chosen polarisation energies ($E_p = 200$, 300, 400 and 500 meV). Note that a source current of $I_0 \sim 5$ nA is transmitted by $\eta \approx 100\%$ for $\alpha_p = \alpha_s$



Figure 6 : Probe diameter d_p and correlated probe current as a function of the convergence angle α_p for our ion optical design [5]. The minimum of d_p ~ 1 nm occurs at an convergence angle of α_p ~ 0.1 mrad with I_p ~ 0.3 pA