Thermal response of a Knudsen-type effusion source to sudden heating-power changes

A. Zehe[1],
Benemérita Universidad Autónoma de Puebla,
Facultad de Cs. Físico-Matemáticas,
Apdo. Post. # 1505, 72000 Puebla, Pue., México.

A Ramírez[2]
Instituto de Ciencias
17 oriente # 1603, Puebla, Pue., México

B. W. Müller
Zentralinstitut für Elektronenphysik, Berlin.

Effusion cells are the basis of nearly all beam sources used in condensed phase molecular beam epitaxy (MBE), although laser-radiation and electron-beam heated sources have also been successfully applied in the growth of certain materials. The temperature of KNUDSEN-type effusion sources is usually adjusted by radiation from a resistance-heated source. The heater is conventionally a refractory metal wire (Tantal) wound noninductively either spirally around the crucible or from end to end supported inside insulating tubing. A thermocouple is used to provide temperature control, and must be placed in a position, e.g. the base of the crucible, in order to provide for a realistic measurement of the cell temperature. The precise control of evaporation and growth rates requires a temperature stability of not less then 0.1% at a base temperature of 1000 °C. This high standard makes a detailed knowledge of the thermal properties of the effusion sources indispensable.

The relation between heating power and source temperature characterizes the stationary behavior, where the use of radiation shields allows to reduce considerably the electrical input. The temperature transition function for a step increase in heating power displays the most important information about the dynamical behavior of the source.

In the present paper an experimental study and model calculations of the response of KNUDSEN-type effusion sources to a step-like change of the heating-power are carried out. It is found, that for small temperature differences the transition function follows approximately an exponential law after some constant delay time has passed. Heat radiation and heat conduction, including of the evaporating material, form the main contributions to the time constant involved.

Keywords: Effusion; Knudsen cells; Temperature behavior

1. Introduction

Effusion cells are the most crucial component of molecular beam epitaxy (MBE) systems. They are used in ultra-high vacuum (UHV) environment to generate ultrapure molecular and atomic beams from a large variety of elements and components [1-4]. The existing great number of different types of effusion cells are specifically designed for the materials to be evaporated. Their operating temperatures might range from –50 °C for organic materials up to 2300 °C for carbon or atomic hydrogen. In MBE, thin films are grown on substrates, placed in a UHV chamber with direct line of sight to several elemental species, each of which is in an evaporation furnace [5, 6]. Through the use of shutters and precise control of the effusion cell temperature almost any material composition and doping can be achieved even with a precision of virtually one atomic layer. The technique is based on a molecular gas flux described first by KNUDSEN almost a century ago [7], which has led to give evaporation furnaces, isothermal evaporation vessels, beam generators or effusion cells with the characteristics of a molecular particle flux the generic name KNUDSEN cells. These cell are most commonly (cylindrical) crucibles with a small knife-edge shaped orifice of about 0.5-1.5 mm diameter in the lid. Effusion through the orifice generates a molecular beam, which spreads out in isotropic distribution following a cosine law.

While the effusion cell design is determined by physical principles of beam generation, the material from which cells are made is important for purity and dynamic behavior.

The particle flux is usually stabilized by a precise temperature control. But there is no way to change the temperature of the evaporating material without delay.

We study in the present paper the dynamic temperature behavior of a KNUDSEN-cell, following sudden changes of the heating power supply.

[1]*e-mail: azehe@prodigy.net.mx*
[2]*e-mail: eduardors@prodigy.net.mx. Tel. 229-55-00 ext. 7851*

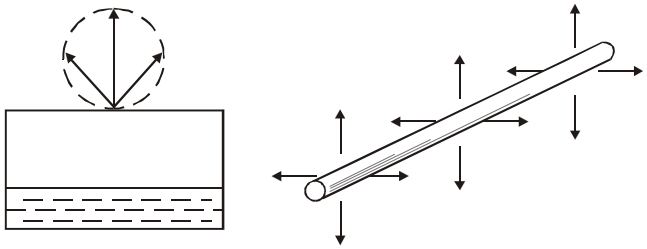

Figure 1. The two extrema of vacuum evaporation: a) ideal Knudsen-evaporation (effusion through an orifice); b) Langmuir-evaporation (vaporizing wire).

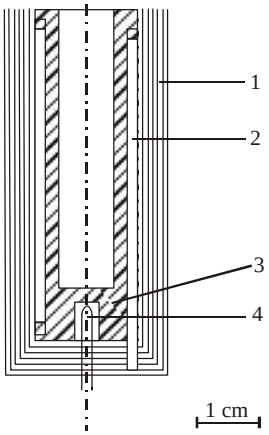

Figure 2. MBE-source. (1) cylindrical radiation shields made of thin tantalum foils; (2) heater (tungsten) in alumina tubes; (3) crucible made of high purity graphite or vitreous carbon; (4) W5%Re-W20%Re thermo-couple.

2. Theoretical considerations

2.1 Temperature dependence of the particle flux

The two extrema of thermal evaporation are set by the ideal Knudsen- and by the Langmuir- evaporation, as illustrated in Figure 1 a and b, respectively.

Langmuir-evaporation means that there are no interactions between the vapour particles and the liquid or solid material (vaporizing wire). On the other hand, the Knudsen- evaporation is characterized by strong interaction of the vapour, liquid and solid phase (equilibrium conditions), and only a small amount of the vapour leaves the reservoir through an orifice without any noticeable disturbance to the equilibrium.

In practice the evaporation is situated between these two extrema. Since the Knudsen-mode can be easily described by means of the kinetic gas theory, this approach is used for the following considerations.

The beam flux distribution can be written as

$$j(\vartheta, T) = j_o(T) f(\vartheta), \qquad (1)$$

where $j_o(T)$ is the particle flux density on the beam axis and $f(\vartheta)$ describes the normalized angular distribution. Since j is given by

$$j = n\bar{v}/4 \qquad (2)$$

(n particle density, $\bar{v}$ mean particle velocity),

$$\bar{v} = \sqrt{8kT/pm} \qquad (3)$$

(T temperature, k Boltzmann-constant, m particle mass), and using the ideal gas law we get

$$j = p/\sqrt{2\delta mkT}\ , \qquad (4)$$

The equilibrium vapour pressure p follows the exponential law

$$p = p_0 \exp(-\Delta H/RT) \qquad (5)$$

(DH enthalpy of evaporation; R universal gas constant). Due to the proportionality

$$j \sim p/\sqrt{T} \qquad (6)$$

one obtains

$$dj/j = [(\Delta H/RT) - ½\ ]dT/T = \alpha \cdot dT/T. \qquad (7)$$

Normally the factor a ranges from 10 to 60 [8], so a temperature variation by 1 K at 1000 K (1‰) results in a flux alteration of 1 to 6 %. This indicates that temperature deviations must be smaller than 1‰ in practical application.

2.2 Radiation shields

Only a small amount of the electric heating power is used to stimulate the evaporation process, which means, that nearly all energy leaves the source by heat radiation.

The evaporation of aluminium, e.g., requires a power of 0.3 W under typical MBE conditions. (This corresponds to a growth rate of about 1 monolayer per second).

Considering a non-shielded source with a surface area of 20 cm^2 at 1500 K, a power of about 400 W will be emitted. Thus, less than 1% of the heating power contributes to the evaporation.

The power input can be considerably reduced by means of radiation shields. In common practice these shields are a set of coaxially arranged and isentropely separated metal foils.

2.3. Temperature dependence of the time constant

For an optimum temperature control a detailed knowledge of both the stationary and the time dependent thermal behaviour of the source is necessary. All parts of the source possess a heat capacity and a limited heat conductivity. Consequently, after the heating power or the radiative conditions (as e.g. by shutter motion [10]) are changed, it will take some time before a new equilibrium is

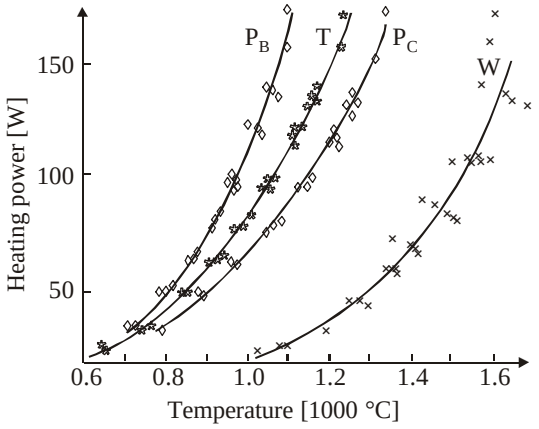

Figure 3. Relationship between the electrical power (required for source heating) and the temperature at four different positions within the source. (P_B) pyrometrically measured temperature at the brim of the crucible; (T) temperature measured with the thermocouple at the bottom of the crucible; (P_C) pyrometrically measured temperature inside the crucible; (W) temperature of the heating wire determined by its resistivity.

established. The term to characterize the transition between two stationary states is called jump response.

Let us consider as a first step, a body of infinite heat conductivity with the temperature T inserted into a radiation field of a constant temperature T_0. According to the Stefan-Boltzmann law the heat transfer dQ/dt is written

$$dQ/dt = \varepsilon\sigma A(T^4 - T_0^4). \quad (8)$$

For small temperature differences of $DT = T - T_0$ the following approximation holds:

$$T^4 - T_0^4 = 4\,T_0^3\,(T - T_0), \quad (9)$$

and therefore the heat transfer becomes

$$dQ/dt = 4\varepsilon\sigma\, A\, T_0^3\,(T - T_0). \quad (10)$$

Replacing dQ *by* CdT (where C is the cells heat capacity) it follows

$$dT/dt = g_r\,(T - T_0) \quad (11)$$

with the reciprocal time constant for radiation g_r,

$$\gamma_r = 4\varepsilon\sigma\, A\, T_0^3/C. \quad (12)$$

The solution of the differential equation (11) is

$$T(t) = T_0 + (T_\infty - T_0)\,(1 - \exp(-\gamma_r t)) \quad (13)$$

(T_∞ source temperature at the new stationary conditions). Note that g_r depends strongly on the temperature as

$$\gamma_r \sim T_0^3. \quad (14)$$

As a second step, we remove the supposition of an infinite heat conductivity by a finite one. Although the heat conduction from the source to the UHV-chambers is negligible, it takes considerable values inside the source. Considering a model similar to that mentioned above the energy transfer by heat conduction is given by

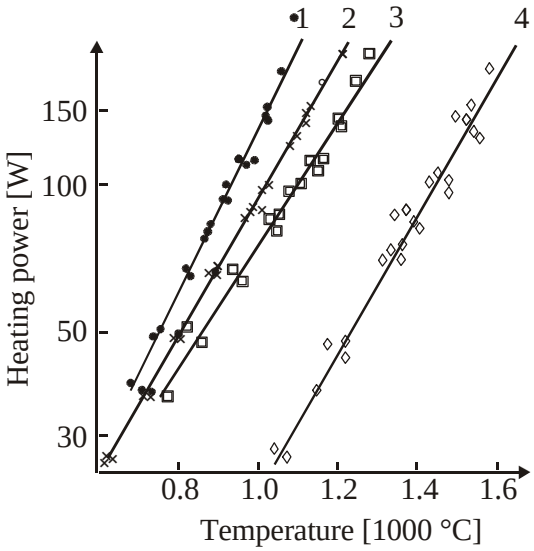

Figure 4. Logarithmic plot of the relation between heating power and source temperature at four positions, as defined in the previous figure.

$$dQ/dt = g\lambda(T - T_0) \quad (15)$$

(g geometry factor, l heat conductivity).

If we again substitute dQ by CdT as in eq. 10, and solve eq. (15), we obtain

$$T(t) = T_0 + ((T_\mu - T_0)[1 - \exp(-\gamma_c t)] \quad (16)$$

with the reciprocal time constant γ_c for heat conduction

$$\gamma_c = g\lambda/C. \quad (17)$$

Eq. (16) displays the same structure as eq. (13), but the finite heat-conduction time constant γ_c is not explicitly temperature dependent. An implicit dependence arises from the temperature dependence of the heat conductivity and the heat capacity. This effect is however small.

Consequently, we can separate the radiation and conduction contributions to the heat transfer if only small temperature changes are considered, i.e. the relation

$$|(T - T_0)/T_0| \ll 1 \quad (18)$$

must be fulfilled.

3. Experimental study

3.1. The thermal effusion source

The MBE-source used for the experiments is shown schematically in Figure 2. The crucible (3) made of high purity graphite and vitreous carbon, respectively, is indirectly heated by a tungsten wire (2) (0.2 mm in diameter, length about 1.1 m) separated by alumina tubes. The crucible and the heater are surrounded by seven radiation shields (1) made of thin tantalum foils. A W5%Re-W20%Re thermocouple (4) is placed in a hole at the bottom of the crucible.

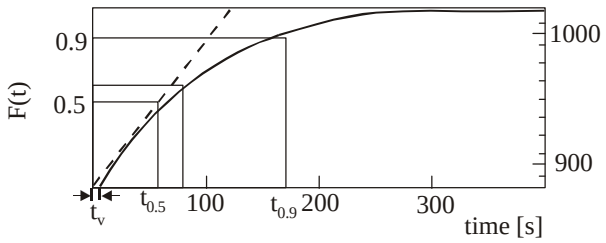

Figure 5: Source response as a function to a step increase in heating power from 63 W to 77 W..

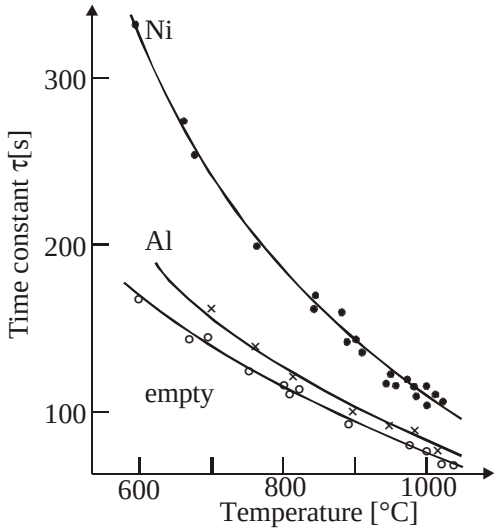

Figure 6: Experimental relation between the time constant τ and the source temperature for the two extrema –the empty cell and the totally filled source, either with Ni or Al.

3.2 Relation between source temperature and heating power

One of the most involved requirements is the determination of the source temperature. In order to get to the temperature distribution of the source, the temperature of the brim and inside the crucible (P_B) and (P_C), respectively, was measured pyrometrically, and the temperature at the bottom was measured by a thermocouple (*T*) (cp. Fig. 3). Furthermore the temperature of the heating wire (*W*) was determined by its resistivity behaviour by means of published data [9]. These measurements show that differences up to 400 K between the crucible and the heater may occur.

It is worth mentioning that the temperature inhomogeneity can be reduced by filling up the crucible. This effect is more pronounced for liquids than for sublimating solids.

A useful feature to characterize the source is the temperature vs. power plot (cp. Figs. 3 and 4). The logarithmically plotted data in Figure 4 demonstrate that above 600 °C, -by comparing the rise of the curves with the value expected from the Stefan-Boltzmann law, the heat from the source is mainly emitted by radiation.

The slope of the curve for the heating wire is steeper because the emissivity of tungsten increases as the temperature in the considered range grows.

3.3. Optimum radiation shielding of the effusion source

The used source without shielding needs a heating power of 123 W to achieve a temperature of 1000 K. Both the top and the bottom emit 8 W each, while 107 W leave the cylinder in radial direction. After a shielding according to Figure 2, the power was reduced to 31 W. It is worth noting that 13% of the surface area (top and bottom) emit more than 50% of the energy. These considerations show that the bottom of the source should be shielded, too.

The top of the source (the orifice) can of course not be shielded, but it is necessary to look for a compromise on the area size of the orifice: on the one hand the orifice should be as small as possible to minimize the emission and, on the other hand, as large as possible in order to minimize the source temperature for a given evaporation rate. The latter fact is important since the amount of contaminations produced by the source increases by an exponential law with the temperature.

The preferred material to build the shields are thin polished foils of tantalum, since tantalum has a low emissivity and is UHV-compatible in an excellent fashion. The tantalum shields reduce not only the energy input into the chamber but improve also the vacuum conditions because tantalum can adsorb gases such as H_2, O_2, N_2 and CO_2 very well.

3.4. The response of the source temperature to a step increase in heating power

In the following, the behaviour of the source temperature by a step increase in heating power from P_1 to P_2 is analyzed. The corresponding temperatures under stationary conditions are called T_1 and T_2, respectively. The transition function *F(t)* is defined by

$$F(t) = (T(t) - T_1)/(T_2 - T_1) \qquad (19)$$

A typical transition function obtained from an empty source is shown in Figure 5. Note that the thermal behaviour of such a MBE-source is comparable to other thermal systems, e.g. contact thermometers.

As shown in chapter 2.3, an exponential law, like $F(t) = 1-\exp(-gt)$ seems a good approximation of the two considered physical processes of thermal conduction and radiation. Nevertheless requires a better fit to the experimentally drawn data (cp. Fig. 5) the inclusion of a short delay time t_v at the beginning of the power step, which was not previously included in the calculus. This delay time is an effect of our lateral experimental procedure and would vanish, if time counting would start at the foot point of the power step function acting on the effusion source. Contrary to t_v the time constant τ is not only temperature dependent (cp. chapter 2.3.) but depends on the heat capacity and the thermal resistance of the

evaporation material, too. In order to estimate both the temperature dependence and the influence on the heat capacity, an empty as well as a niquel- source (Ni) was investigated (cp. Fig. 6).

We have chosen Ni because of its very high heat capacity per volume. For a source filled with aluminium (Al), typical in MBE, the time constant τ is situated between these two extrema.

4. Summary

In most cases of molecular beam deposition, reproducible and stable particle fluxes realized by a precise temperature control of the effusion ovens are highly important. This is especially the case when well defined doping concentrations and mole fractions in complex compounds are needed. An efficient temperature control requires data on both the steady-states and dynamical thermal behaviour of the source.

The relationship between heating power and cell temperature is an expression, which characterizes the steady-state behaviour of the sources. Since the heating power is always related to unintentional gas emission into the UHV-system by an exponential law, the power input should be as low as possible. The heating power, in order to produce a certain source temperature, can considerably be reduced by the application of radiation shields. The efficiency of the shield ensemble depends on the number of shields, on their emissivity and arrangement.

The temperature transition function for a step increase in heating power furnishes the most important information on the dynamical behaviour of the source. It has been shown that for small temperature differences the transition function can be approximated by an exponential law, if a constant delay time is taken into account. The time constant τ used for this law consists of a contribution related to the heat radiation (proportional to T^3) and a nearly temperature independent contribution derived from the heat conduction.

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