# Proposal for new experimental schemes to realize the Avogadro constant.

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## Abstract

We propose two experimental schemes to determine and so to realize the Avogadro constant $N_A$ at the level of $10^{-7}$ or better with a watt balance experiment and a cold atom experiment measuring $h/m(X)$ (where $h$ is the Planck constant and $m(X)$ the mass of the atom $X$). We give some prospects about achievable uncertainties and we discuss the opportunity to test the existence of possible unknown correction factors for the Josephson effect and quantum Hall effect.



## 1. Introduction

The best estimates of the fundamental constants are determined by the Codata adjustments [1, 2]. A key weakness of this adjustment is the lack of redundancy of input data. Especially, this makes the determination of the Planck constant $h$ and the Avogadro constant $N_A$ less confident [2]. In this paper, we propose to associate watt balance and cold atoms experiments to define new direct experimental ways to realize the Avogadro constant with a competitive uncertainty in comparison with the best determination obtained from the molar volume of a silicon crystal ($3.1\times10^{-7}$) [3]. Moreover, the recent proposal of redefinition of the kilogramme either by fixing the value of $h$ or the value of $N_A$ [4, 5, 6] will be reinforced by many independent experimental determinations or comparisons of these constants. Associated with a determination of $N_A$ at the level of $10^{-8}$, the proposed experiments can be used to check the validity of the product $K_J^2 R_K$ (where $K_J$ and $R_K$ , the Josephson and the von Klitzing constants, are respectively associated to the Josephson and the quantum Hall effects).

## 2. Principle of the $N_A$ realization

### 2.1 Quantities measured by the two experiments

From its definition, $N_A$ can be expressed as the ratio between atomic and macroscopic quantities such as the atomic mass and the molar mass of any element. As it is already done in the above mentioned single crystal silicon determination, any couple of experiments giving access to these quantities may be considered. Another possible combination consists in bringing together a watt balance intended to link the kilogram to an invariant quantity and a $h/m(X)$ experiment. Indeed, integrating these two experiments leads to the determination of $N_A$ using the relation :

$$N_A = \frac{1}{h}\frac{h}{m(X)}A_r(X)M_u \tag{1}$$

where $A_r$(X) is the relative atomic mass of $X$ and $M_u$ is the molar mass constant ($M_u$=$10^{-3}$kg mol$^{-1}$).

The watt balance experiment consists in comparing a mechanical power to an electromagnetic power [7]. This comparison is performed in two steps. In a static phase, the Laplace force on a coil driven by a DC current and submitted to an induction field is compared to the weight of a standard mass, linked to the kilogram $M$. In a dynamic measurement, the voltage induced at the terminals of the same coil is measured when it is moved in the same field at a known velocity $V$. The measurement of electrical quantities by comparison to the Josephson effect and the quantum Hall effect (QHE) allows then to link the mass of the kilogram to the product $K_J^2 R_K$.

$$MK_J^2R_K = \frac{A}{g^{(w)}V} \tag{2}$$

where $A = \frac{f_1 f_2}{p}$ is proportional to the product of the two Josephson frequencies involved in the voltage measurements during the static and dynamic phases [8]. The dimensionless $p$ is relative to the calibration of a resistance standard against the quantum Hall effect and $g^{(w)}$ is the local acceleration seen by the macroscopic mass $M$. Writing the quantities to be measured in the experiment between brackets {}, the watt balance can determine $\{K_J^2R_K\}$ if $g^{(w)}$ is measured independently with an absolute gravimeter, as well as $\{K_J^2R_Kg^{(w)}\}$.

The ratio $h/m(X)$ is determined by measuring the recoil velocity ($v_r = \hbar k/m(X)$) defined as the velocity induced by light when an atom at rest absorbs a photon of momentum $\hbar k$. The ratio $h/m(^{133}Cs)$ has been measured for the first time, at Standford, using an atom interferometer with a relative uncertainty of 15 ppb [9]. In another experiment, in Paris, we have measured the ratio $h/m(^{87}Rb)$ with a relative uncertainty of 13 ppb using Bloch oscillations in an optical lattice [10]. A narrow velocity class is selected from cold atoms sample with a Raman -pulse. This velocity class is accelerated with Bloch oscillations. This process allows us to transfer efficiently a high number of photon momenta [11]. The final velocity is measured with another Raman pulse. This experiment can run in two modes leading to two ways to determine the Avogadro constant labelled $N_A^{(1)}$ and $N_A^{(2)}$ hereafter.

In the first mode, a vertical optical standing wave is used to hold the atoms against gravity [12]. The atoms oscillate at the same place at the Bloch frequency ($\nu_{Bloch}$):

$$\nu_{Bloch} = \frac{m(^{87}Rb)g^{(a)}\lambda_{opt}}{2h} \tag{3}$$

where $\lambda_{opt}$ is the wavelength of the optical wave and $g^{(a)}$ is the local acceleration seen by the atoms. The quantity $\{h/m(^{87}Rb)g^{(a)}\}$ is measured in terms of frequencies.

If the two experiments are brought close enough, the two local accelerations $g^{(w)}$ and $g^{(a)}$ can be compared accurately with relative gravimeters [13]. Combining the quantities measured by the two experiments leads to determine:

$$\{K_J^2R_Kg^{(w)}\}\{\frac{h}{m(^{87}Rb)g^{(a)}}\}\{\frac{g^{(a)})}{g^{(w)}}\} \tag{4}$$

In the second mode of the $h/m(^{87}Rb)$ experiment, the atoms are accelerated up and down with Bloch oscillations. The resulting differential measurement of $h/m^{87}Rb$ is independent of the local acceleration $g$ [10]. Again the quantity $\{h/m(^{87}Rb)\}$ is measured in terms of frequencies. The combination of the two experiments gives:

$$\{K_J^2 R_K\}\{\frac{h}{m(^{87}Rb)}\} \tag{5}$$

## 2.2 Determination of NA

The values assigned by theory to $K_J$ and $R_K$ are :

$$K_J = \frac{2e}{h} \tag{6}$$

and

$$R_K = \frac{h}{e^2} \tag{7}$$

where $e$ is the elementary charge. The theoretical value of $K_J^2 R_K$ is then $4/h$.

If these relations are considered to be exact, the realization of $N_A$ labelled $N_A^{(1)}$ and $N_A^{(2)}$ can be written as :

$$N_A^{(1)} = \{\frac{K_J^2 R_K g^{(w)}}{4}\}\{\frac{h}{m(^{87}Rb)g^{(a)}}\}\{\frac{g^{(a)})}{g^{(w)}}\}A_r(^{87}Rb)M_u \tag{8}$$

$$N_A^{(2)} = \{\frac{K_J^2 R_K}{4}\}\{\frac{h}{m(^{87}Rb)}\}A_r(^{87}Rb)M_u \tag{9}$$

Notice that the realization of $N_A^{(1)}$ does not need the knowledge of the absolute values of the local gravity $g^{(a)}$ and $g^{(w)}$ but only their relative values. In the case of $N_A^{(2)}$, the knowledge of requires an absolute measurement of $g^{(w)}$ (see eq. 2).

## 2.3 Test of $K_J^2 R_K$

However, proposing a new definition of the kilogram in terms of a fundamental constant requires to lay on both theoretical and experimental arguments. Even if the reproducibility of the quantum Hall effect and the Josephson effect has been tested with a relative uncertainty better than $1\times 10^{-10}$ under various experimental conditions (material, temperature,...), there is no experimental proof at this level that $K_J$ and $R_K$ are equal to their theoretical values [14, 15]. At present, the only way to verify that $R_K$ is effectively equal to $h/e^2$ consists in comparing its value obtained from an experiment involving a QHE setup and the Lampard calculable capacitor [16] to those derived from other experiments, such as determinations. The situation is similar for $K_J$ (assumed to be equal to $2e/h$) whose experimental knowledge is issued, up to now, from the use of electrometers [17], even if a determination can be deduced from the watt balance experiment, provided resistance measurements are made in SI values by comparison, for example to a Lampard calculable capacitor.

A test of this exactness has been done in the last Codata adjustment, using the multivariate analysis. The inconsistencies observed among certain input data have conducted to relax the strict condition of equality between $R_K$ and $K_J$ and their theoretical values. Two more adjusted constants $\epsilon_J$ and $\epsilon_K$ describing unknown correction factors have been added in the adjustment. The expressions of $K_J$ and $R_K$ then become:

$$K_J = \frac{2e}{h}(1+\epsilon_J) \tag{10}$$

and

$$R_K = \frac{h}{e^2}(1+\epsilon_K) \tag{11}$$

Therefore the relations issued from the two approaches proposed for the experiment may be rewritten as:

$$N_A^{(1)} = \{\frac{K_J^2 R_K g^{(w)}}{4}\}\{\frac{h}{m(^{87}Rb)g^{(a)}}\}\{\frac{g^{(a)})}{g^{(w)}}\}\frac{A_r(^{87}Rb)M_u}{(1+\epsilon_J)^2(1+\epsilon_K)} \tag{12}$$

$$N_A^{(2)} = \{\frac{K_J^2 R_K}{4}\}\{\frac{h}{m(^{87}Rb)}\}\frac{A_r(^{87}Rb)M_u}{(1+\epsilon_J)^2(1+\epsilon_K)} \tag{13}$$

# 3. Discussion

We introduce in this paragraph the present status of the different uncertainties of these two possible realizations.

For the determination of $N_A^{(1)}$, the overall relative standard uncertainty is presently limited by the uncertainty of the quantity $\{\frac{h}{m(^{87}Rb)g^{(a)}}\}$. This ratio has been measured in a preliminary experiment with a relative uncertainty of about $10^{-6}$ [12], mainly due to vibrations and collisions with the background vapor. These two technical limitations can be overcome by using a more suitable vacuum chamber where the Bloch oscillations take place and by improving the vibration isolation. For example, 4000 Bloch oscillations have been recently observed during 10s in the gravity field [18]. The other components of uncertainty are smaller : the uncertainty of the quantity $\{K_J^2 R_K g^{(w)}\}$ can be extrapolated from [19] at the level of $4\times 10^{-8}$, the relative atomic mass of rubidium $Ar(^{87}Rb)$ is known with an uncertainty better than $2\times 10^{-10}$ [20] and the gravity transfer can be performed with an uncertainty of the order $1\times10^{-9}$ if the two experimental setups are close enough [REFERENCE ?].

The different contributions to the relative standard uncertainty ($u_r$) of $N_A^{(2)}$ extrapolated from the different results are listed in the following table. Different values of $\epsilon_J$ and $\epsilon_K$, taking (or not) into account some input data, are given in [2]. We use here the values $\epsilon_J$ and $\epsilon_K$ calculated with all the input data :

| Quantity | Value(uncertainty) | Ref. |
|---|---|---|
| $[h/m_{Rb}]$ | 4.591 359 291 (61) $10^{-9}\mathrm{m^2s^{-1}}$ | [10] |
| $4/[K_J^2 R_K]$ | 6.626 069 01(34) $10^{-34}$ Js | [19] |
| $A_r(^{87}Rb)$ | 86.909 180 520 (15)u | [20] |
| $\epsilon_J$ | -126 (81) $10^{-9}$ | [2] |
| $\epsilon_K$ | 23 (19) $10^{-9}$ | [2] |

Table 1

If it is assumed that there is no statistical significant evidence that the basic relations for $K_J$ and $R_K$ are not exact [2], $\epsilon_J$ and $\epsilon_K$ as well as their uncertainties can be considered as equal to 0. Then, the relative uncertainty on $N_A^{(2)}$ is at the level of $5.3\times 10^{-8}$.

$$N_A^{(2)} = 6.022\ 141\ 83\ (33)\ 10^{23}\ \mathrm{mol}^{-1}$$

This values may be compared to the one issued from the silicium [3] and to the one recommended by Codata [2]:

$$N_A^{(Si)} = 6.022\ 135\ 3\ (18) 10^{23}\ \mathrm{mol}^{-1}$$
$$N_A^{(Codata)} = 6.022\ 141\ 5\ (10)\ 10^{23}\ \mathrm{mol}^{-1}$$

If we now consider the possible values of $\epsilon_J$ and $\epsilon_K$, and the associated uncertainties issued from the Cotata tests (see table 1), a new value of $N_A^{(2)}$ can be determined :

$$N_A^{(2)} = 6.022\ 143\ 21\ (103)\ 10^{23}\ \mathrm{mol}^{-1}$$

The covariance factor between $\epsilon_J$ and $\epsilon_K$ is extremely small as $\epsilon_K$ is determined mainly by the measurements of and $R_K$, while $\epsilon_J$ should depend only weakly on these measurements [21]. Taking into account a null value for this covariance leads to a relative uncertainty on $N_A^{(2)}$ of $2.8 \times 10^{-7}$.

If, as mentioned by the groups in charge of the silicon project, an uncertainty of $2 \times 10^{-8}$ is expected in the future [22], the proposed determinations conduct to establish a direct link between $R_K$ and $K_J$ derived from solid state physics and $h$ derived from atomic physics. In that case, gathering the two experiences could improve significantly our confidence in the coherence of the phenomena on which a new definition of the kilogram could be established and on the experimental data on which its mise en pratique could be based. Considering the above mentioned uncertainties, this could lead to know the product $hK_J^2R_K/4$ with a relative uncertainty of $5.3 \times 10^{-8}$.

# 4. Conclusion

We propose here new competitive schemes to realize the Avogadro constant based on the conjunction of $h/m(^{87}Rb)$ experiment and watt balance experiment where all quantities are measured in terms of frequency. This proposal emphasizes the strong interest of having a cold atom experiment nearby the watt balance. The versatility of a cold atom experiment which can be used to measure either $g^{(w)}$ [23] or $h/m(^{87}Rb)$ enables the realization of $h$ and $N_A$ with the same watt balance. Presently, provided it is considered that $K_J$ and $R_K$ are equal to their theoretical value without any uncertainty, $N_A$ can be realized at a level of $5.3 \times 10^{-8}$. This uncertainty rises to $2.8 \times\ 10^{-7}$ if one take into account possible correction factors discussed by Codata 2002. This shows that the knowledge on the von Klitzing and Josephson constants may have a great influence on the numerical values of fundamental constants and that any experiment to improve this knowledge must be encouraged. The aim of the new experiment using an enriched silicon sphere is to reach a $2 \times 10^{-8}$ relative uncertainty for the determination of $N_A$. An agreement at this level of uncertainty with the values of $N_A$ issued from the scheme proposed in this paper could then lead to a test of the equality of the product $K_J^2R_K$ with $4/h$ with a relative uncertainty of $5.7 \times 10^{-8}$. This strongly emphasizes the interest of such determinations before the redefinition of the kilogram and its *mise en pratique*.

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