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# Anomalous cooling of the parallel velocity in seeded beams

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We have measured the parallel velocity distribution of a lithium supersonic beam produced by seeding lithium in argon. The parallel temperature for lithium is considerably lower than the calculated parallel temperature of the argon carrier gas. We have extended the theory of supersonic cooling to calculate the parallel temperature of the seeded gas, in the limit of high dilution. The theoretical result thus obtained is in good agreement with our observations.

## I. INTRODUCTION

We have observed an unexpected cooling effect in an argon supersonic beam seeded with a small amount of lithium: the lithium parallel temperature is smaller than the argon parallel temperature. This effect is surprisingly large, the lithium parallel temperature being 4 times smaller than the calculated argon parallel temperature, whereas the common assumption is that these two parallel temperatures are roughly equal.

Although surprising, this effect can be explained by an extension of the theory giving the terminal parallel temperature in supersonic beams of pure gases. This theory, first developed by Anderson and Fenn in 1965 [1], has been completed by Toennies and Winkelmann [2] in 1977 and Beijerinck and Verster [3] in 1981 (for a brief review, see ref. [2]). In his 1988 review on free jet sources, D.R. Miller [4] discusses the case of expansion of binary mixtures: this case has been studied by various authors but their results, which are most accurate for a dilute heavy species, do not predict a large temperature difference in our case.

The basic phenomenon is due to the exchange of energy between the alkali atoms and the rare gas atoms. Two effects are important: i) the alkali-rare gas interaction has a longer range than the rare gas-rare gas interactions so that the expansion cooling goes on for a longer time for the alkali atoms; ii) the mass ratio plays an important role in the energy exchange between the two species and, as shown below, the parallel temperature of the seeded gas (mass  $m_2$ ) is considerably lower than the parallel temperature of the carrier gas (mass  $m_1$ ) when  $m_2 \ll m_1$ .

In this paper, we first describe our measurement of the parallel temperature of lithium seeded in argon. Then, we recall the theory explaining the final temperature in supersonic beams and we describe its extension to the case of a gas mixture in the limit of a high dilution. We thus get differential equations coupling the parallel and

perpendicular temperatures of the seeded gas to the same quantities of the carrier gas. Finally, we compare our theoretical result to our experiment as well as to another evidence of this anomalous cooling effect.

## II. OUR MEASUREMENT

We have measured the parallel temperature of our lithium beam by laser induced fluorescence. The fluorescence of the  ${}^7\text{Li}$  isotope (92.5% natural abundance) is excited by a single frequency laser operating at 671 nm, using the  ${}^2S_{1/2} - {}^2P_{3/2}$  resonance transition. Two laser beams are used, one near normal incidence measures the transverse velocity distribution, while the other one, working with an incidence close to  $45^\circ$  is sensitive to a combination of the parallel and perpendicular velocity distributions. We use a low laser power density in order to prevent any saturation broadening. Figure 1 presents a typical spectrum of the fluorescence as a function of the laser frequency. Each laser beam excites two resonance lines corresponding to the two hyperfine levels  $F = 1$  and  $F = 2$  of the  ${}^2S_{1/2}$  ground state, with an hyperfine splitting equal to 803.5 MHz, while the very small hyperfine structure of the  ${}^2P_{3/2}$  excited state cannot be resolved [5].

The distance between the lines corresponding to the same transition excited by the two laser beams gives access to the mean velocity of the beam, while the linewidths give access to the widths of the velocity distributions. The observed perpendicular velocity distribution depends on the width of the observed part of the lithium beam and our observation corresponds to a perpendicular temperature of 0.4 K. As expected, the parallel velocity distribution is broader than the perpendicular velocity distribution (in our excitation geometry with a  $45^\circ$  incidence, the Doppler width is only 0.71 times the width we would get if the laser and atomic beams were parallel). The measured parallel temperature is  $T_{\parallel} = 6.6$  K, while the calculated parallel temperature of our argon beam is 26 K.

## III. TERMINAL PARALLEL TEMPERATURE IN SUPERSONIC EXPANSIONS

We consider here only the case of monoatomic gases. The starting point is the Boltzmann equation. In the

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frame moving with the hydrodynamic velocity  $u$ , the velocity distribution is assumed to be Maxwellian with different parallel and perpendicular temperatures  $T_{\parallel 1}$  and  $T_{\perp 1}$  :

$$f_1(\mathbf{v}) = \left( \frac{m_1}{2\pi k_B T_{\parallel 1}} \right)^{1/2} \times \frac{m_1}{2\pi k_B T_{\perp 1}} \times \exp \left[ -\frac{m_1 v_{\parallel}^2}{2k_B T_{\parallel 1}} - \frac{m_1 v_{\perp}^2}{2k_B T_{\perp 1}} \right] \quad (1)$$

$m_1$  is the atomic mass. We use local cartesian coordinates, the  $x$  and  $y$  axis being in the perpendicular directions and the  $z$  axis being in the parallel direction. The theoretical treatment of Toennies and Winkelmann [2] (noted below TW) and the one of Beijerinck and Verster [3] (noted below BV) are different but equations (14),(15b) and (16) of reference [2] are equivalent to equations (29a,b) and (B6,7) of reference [3]. However, small terms of the order of  $S_{\parallel}^{-2}$ , where the parallel speed ratio  $S_{\parallel}$  is defined by  $S_{\parallel} = \sqrt{mu^2/(2kT_{\parallel})}$  are neglected in the BV calculations, which are a good approximation only when the terminal value  $S_{\parallel\infty}$  of the parallel speed ratio is large. As we are interested in this case here, we follow the BV calculations because they are simpler. The parallel and perpendicular temperatures are given by:

$$\frac{dT_{\perp 1}}{dz} = -\frac{2T_{\perp 1}}{z} + F \text{ and } \frac{dT_{\parallel 1}}{dz} = -2F \quad (2)$$

where  $F$  is due to the collision-induced energy transfer from parallel to perpendicular degrees of freedom:

$$F = \frac{n_1(z)}{2k_B u} \int g \frac{d\sigma_{1,1}(g)}{d\Omega} \Delta E f_1(\mathbf{v}_1) f_1(\mathbf{v}_2) d^3 \mathbf{v}_1 d^3 \mathbf{v}_2 d\Omega \quad (3)$$

$\mathbf{v}_1$  and  $\mathbf{v}_2$  are the atom velocities before the collision and  $g = |\mathbf{v}_1 - \mathbf{v}_2|$  is their relative velocity. The indices 1, 1 recall that the collision considered here involve two atoms of species 1.  $\Delta E$  is the energy transferred during one collision from the parallel degree of freedom to the perpendicular ones; after averaging over the azimuth of the final relative velocity around the initial relative velocity,  $\Delta E$  is given by :

$$\Delta E = \frac{m}{8} [g_x^2 + g_y^2 - 2g_z^2] [1 - \cos^2 \chi] \quad (4)$$

where  $\chi$  is the deflection angle.  $F$  is well approximated by a linear function of  $(T_{\parallel 1} - T_{\perp 1})$  :

$$F = \Lambda(z)(T_{\parallel 1} - T_{\perp 1}) \text{ with } \Lambda(z) = 16n_1(z)\Omega_{1,1}^{(2,2)}(T_m)/(15u_{\infty}) \quad (5)$$

Here,  $\Omega_{1,1}^{(l,s)}(T_m)$  defined by reference [6] (see equation 7.4-34) is a thermal average at the mean temperature  $T_m = (T_{\parallel 1} + 2T_{\perp 1})/3$  of the collision integral  $Q_{1,1}^{(l)}$  given by:

$$Q_{1,1}^{(l)} = \int \frac{d\sigma_{1,1}(g)}{d\Omega} (1 - \cos^l \chi) d\Omega \quad (6)$$

When quantum effects can be neglected, if the interaction potential is approximated by a 12-6 Lennard-Jones potential with a  $C_6(1, 1)$  long-range coefficient and if the relative kinetic energy is small with respect to the interaction potential well depth  $\epsilon$ , the  $\Omega_{1,1}^{(2,2)}(T)$  integral is given by [3]:

$$\Omega_{1,1}^{(2,2)}(T) = 2.99 (2k_B T/m_1)^{1/2} (C_6(1, 1)/k_B T)^{1/3} \quad (7)$$

Knowing  $\Lambda(z)$ , which behaves like  $z^{-2}T_m^{1/6}$ , the differential equations (2) can be integrated numerically, thus relating the terminal parallel temperature  $T_{\parallel\infty}$  to the source conditions. The tradition is to give the terminal value  $S_{\parallel\infty}$  of the parallel speed ratio :

$$S_{\parallel\infty} = A \left[ n_0 d (C_6/k_B T_0)^{1/3} \right]^{\alpha} \quad (8)$$

$n_0$  and  $T_0$  are the density and temperature in the source,  $d$  the nozzle diameter. Using SI units, the various values of  $A$  and  $\alpha$  are  $A = 1.413$  and  $\alpha = 0.53$  obtained by TW [2],  $A = 1.313$  and  $\alpha = 0.545$  obtained by BV [3],  $A = 1.40$  and  $\alpha = 0.495$  obtained also by BV from a fit of experimental  $S_{\parallel\infty}$  values for argon expansions.

#### IV. GENERALIZATION TO THE CASE OF A MIXTURE OF TWO MONOATOMIC GASES

Our calculation is made in the case of a high dilution, i.e. when the seeded gas density  $n_2$  is considerably smaller than the carrier gas density  $n_1$ . We neglect the velocity slip i.e. we assume the same hydrodynamic velocity  $u$  for both species. Because  $n_2 \ll n_1$ , the expansion of the carrier gas is not modified by the presence of the seeded gas and the equations written above remain valid. For the seeded gas, we write similar equations, considering only the collisions with atoms of the carrier gas. This calculation is not detailed here because it is more complex. In order to linearize the equations, we have remarked that the products  $gQ_{1,2}^{(l)}(g) \propto g^{1/3}$  vary slowly with  $g$  and we have treated these quantities as constant. Then, with some further approximations, we get the differential equations relating  $T_{\parallel 2}$  and  $T_{\perp 2}$ :

$$\frac{dT_{\perp 2}}{dz} = -\frac{2T_{\perp 2}}{z} + \Lambda(z)\rho_s \frac{m_1}{M} [T_{\parallel av} - T_{\perp av}] - 4\Lambda(z)\rho_s \rho_o \frac{\mu}{M} [T_{\perp 2} - T_{\perp 1}] \quad (9)$$

$$\frac{dT_{\parallel 2}}{dz} = -2\Lambda(z)\rho_s \frac{m_1}{M} [T_{\parallel av} - T_{\perp av}] - 4\Lambda(z)\rho_s \rho_o \frac{\mu}{M} [T_{\parallel 2} - T_{\parallel 1}] \quad (10)$$

with  $T_{\parallel av} = \beta T_{\parallel 1} + \alpha T_{\parallel 2}$ ,  $T_{\perp av} = \beta T_{\perp 1} + \alpha T_{\perp 2}$ ,  $\alpha = m_1/(m_1 + m_2)$  and  $\beta = m_2/(m_1 + m_2)$ .  $\rho_s$  is the ratio of  $\Omega_{i,j}^{(2,2)}$  collision integrals:

$$\rho_s = \frac{\Omega_{1,2}^{(2,2)}}{\Omega_{1,1}^{(2,2)}} = \left[ \frac{C_6(1,2)}{C_6(1,1)} \right]^{1/3} \times \left[ \frac{m_1 + m_2}{2m_2} \right]^{1/2} \quad (11)$$

$\rho_o$  is the ratio of the  $l = 1$  and  $l = 2$  angle-averaged cross-sections  $\rho_o = Q_{1,2}^{(1)}/Q_{1,2}^{(2)}$ . Using the low energy deflection function for a 12-6 Lennard-Jones potential (see appendix A of [3]), we have calculated  $\rho_o = 1.32$ .

Following BV, we have introduced reduced temperatures and a reduced  $z$ -coordinate. Then, the numerical integration of the coupled equations gives the temperatures represented in figure 2. At the end of the expansion, the parallel and perpendicular temperatures of the seeded gas are lower than the similar quantities for the carrier gas. The parallel temperature ratio  $T_{||2}/T_{||1}$  can be substantially lower than 1. The minimum value for temperature  $T_{||2}$  is reached when the right-hand side of equation (9) vanishes. Neglecting the perpendicular temperatures, we thus get:

$$\frac{T_{||2,min}}{T_{||1\infty}} = \frac{m_2\rho_o}{m_1 + 2m_2\rho_o} \quad (12)$$

This limiting value is reached if the number of 1 – 2 collisions is sufficiently larger than the number of 1 – 1 collisions, i.e. if the ratio  $\rho_s$  is large enough. We have plotted in figure 3 the terminal value of the ratio  $T_{||2}/T_{||1}$  for various values of the ratio  $\rho_s$  and for different values of the mass ratio  $m_2/m_1$ .

## V. COMPARISON WITH EXPERIMENTAL RESULTS

We may now compare our theoretical and experimental results for lithium seeded in argon. In our experiment with  $p_0 = 333$  millibar,  $T_0 = 943$  K and a nozzle diameter  $d = 200$   $\mu\text{m}$ , the parallel temperature of lithium is  $T_{||2\infty} = 6.6$  K. Using equation (8) with  $A = 1.40$  and  $\alpha = 0.495$  [3], we deduce the argon parallel temperature from our source conditions  $T_{||1\infty} = 26$  K. The calculated temperature may be overestimated, because the true collision cross-section is underestimated [4] by equation (7) when the source temperature is comparable to or larger than the argon-argon well depth  $\epsilon/k_B \approx 143$  K [7,8]. However, as we are using an empirical law based on experiments with source temperatures of 288 K and 380 K, we think that the overestimation of the argon parallel temperature should not be very large. We thus get a parallel temperature ratio  $T_{||2\infty}/T_{||1\infty} = 0.25$ . This value is larger than the limiting ratio to  $T_{||2,min}/T_{||1\infty} = 0.16$  for lithium in argon and it is smaller than our theoretical result  $T_{||2\infty}/T_{||1\infty} = 0.31$  obtained with  $\rho_s = 2.55$  deduced from the  $C_6$  values of argon-argon and lithium-argon interaction [9]. The difference between theory and experiment may be due partly to the overestimation of the argon parallel temperature discussed above, partly to the approximations done in our theory.

A similar beam with sodium seeded in argon was built by D. Pritchard and coworkers [10,11] with a source temperature close to  $T_0 = 1000$  K, an argon pressure up to  $p_0 = 2000$  Torr and a nozzle diameter  $d = 70$   $\mu\text{m}$ . The measured velocity  $u = 1040 \pm 2$  m/s corresponds to  $T_0 = 1039$  K and the rms velocity width, deduced from the atomic diffraction pattern, was found equal to  $3.7 \pm 0.4\%$ , corresponding to  $T_{||2\infty} = 4.1 \pm 0.9$  K. From the calculated argon parallel temperature  $T_{||1} = 14$  K at the largest pressure, we get the parallel temperature ratio  $T_{||2\infty}/T_{||1\infty} = 0.29 \pm 0.06$ , very close to the limiting ratio  $T_{||2,min}/T_{||1\infty} = 0.30$  for this mass ratio. Using the  $C_6$  value of sodium-argon interaction from reference [9] ( $\rho_s = 1.67$ ), our theoretical result is  $T_{||2\infty}/T_{||1\infty} = 0.61$ , substantially larger than the experimental value. A part of this difference can be explained as in the lithium case but some other effect is probably involved.

## VI. CONCLUSION

In this letter, we have described a measurement of the parallel velocity distribution of a supersonic beam of lithium seeded in argon, with a high dilution. The measured lithium parallel temperature 6.6 K is considerably lower than the calculated parallel temperature of the argon beam near 26 K.

We have extended the theory of the velocity distribution in supersonic beams to a mixture of monoatomic gases, in the high dilution limit. We have established differential equations describing the parallel and perpendicular temperatures of the seeded gas; these equations explain the observed effect as a consequence of several circumstances: role of the mass ratio in energy exchange during collisions of the two species, longer range of the lithium-argon interaction as compared to argon-argon interaction. We have deduced the lowest possible value of the temperature ratio  $T_{||2\infty}/T_{||1\infty}$ . This ratio may be quite low when a light species is seeded in a heavy species with  $m_2 \ll m_1$ .

Comparison of this theory with our experimental results is satisfactory and we have also found evidence of the same effect in data published by D. Pritchard and coworkers on a sodium beam seeded in argon. In both cases, the ratio of the measured alkali parallel temperature to the calculated argon parallel temperature is lower than what our theory predicts. This discrepancy may be partly due to an overestimation of the terminal argon parallel temperatures and we have good reasons to think that such an overestimation may result from our use of Beijerinck and Verster semi-empirical formula in the range of high source temperature where it has not been tested. This discrepancy may also be due to the approximations we have made in our calculation of the parallel temperature of the seeded gas and we are going to test these approximations by more detailed calculations.

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FIG. 1. Laser induced fluorescence signal as a function of the laser frequency: the dots represent the experimental data and the full curves are the Gaussian fits to each line. The Fabry-Perot used for calibration has a free spectral range equal to  $251.4 \pm 0.5$  MHz. The fluorescence peaks are labeled by the ground state  $F$  value ( $F = 1$  or  $2$ ) and by a letter corresponding to the laser beam: A for the beam near  $45^\circ$  incidence, B for the beam near normal incidence.

FIG. 2. The reduced temperatures  $\tau$  are plotted as a function of the reduced  $z$ -coordinate  $\zeta$ . This plot, which is identical to figure 5 of Beijerinck and Verster [3] for  $\tau_{\parallel 1}$  and  $\tau_{\perp 1}$ , shows the anomalous cooling effect for species 2. We have used  $\rho_o = 1.32$  and  $\rho_s = 2.55$  corresponding to lithium seeded in argon.

FIG. 3. The calculated parallel temperature ratio  $T_{\parallel 2\infty}/T_{\parallel 1\infty}$  is plotted as a function of the mass ratio  $m_2/m_1$  (spanning the range from hydrogen to rubidium seeded in argon) for several values of the ratio  $\rho_s$ , with  $\rho_o$  fixed,  $\rho_o = 1.32$ . From bottom to top,  $\rho_s = \infty$ ,  $\rho_s = 1.67$  (sodium case),  $\rho_s = 2.55$  (lithium case). The experimental points corresponding to lithium and sodium seeded in argon are represented by stars.