Atom symmetry breaking in Van der Waals surface interaction: metastable level coupling in Argon and Krypton atoms on a Copper surface

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Because of the cylindrical symmetry around the normal $\hat{z}$ to a planar surface, the van der Waals atom-surface interaction Hamiltonian exhibits a *quadrupolar* component proportional to $D_z^2 - D^2/3$, where $\mathbf{D}$ is the atomic dipole operator, in addition to the scalar term in $4D^2/3$ [1]. The former term is a off-diagonal coupling which breaks the symmetry of the atomic wave functions and mixes energy levels of same parity. For instance in a rare gas atom heavier than He the $^3P_0$ metastable level is coupled to the lower $^3P_2$ metastable level. The $^3P_0 -^3 P_2$ population transfer induced by the quadrupolar van der Waals term can be estimated by use of a simple perturbation treatment. The transition rate is given by: $Q^2/\Delta E$, where ΔE is the $^3P_0 -^3 P_2$ energy splitting and:

$$Q = \frac{1}{16z^3} <^3 P_2|D_z^2|^3P_2 > .$$

Because of the translational invariance along the surface plane the momentum transfer is normal to the surface. For an atom impinging the surface at a grazing angle θ_i the final scattering angle with respect to the plane is θ_f such that:

$$v_i \cos\theta_i = v_f cos\theta_f$$

where v_i is the initial velocity and $v_f = (v_i^2 + 2\Delta E/m)^{1/2}$ is the final one. The energy gain ΔE (174 meV for Ar, 650 meV for Kr) is large compared to a typical kinetic energy in the thermal range ($\approx$ 70 meV). This leads to large deflection angles : about 60^o for argon, 70^o for krypton.

The experimental device is quite simple. $^3P_0$ and $^3P_2$ metastable atoms are produced with a relative ratio 1:5 by a pulsed electronic bombardment of an effusive atom beam. A velocity selection ($\delta v/v \approx 10\%$) is then achieved by passing the beam through a synchronised slotted disk Then the atoms traverse a 100μm-wide and 50μm-thick copper slit. *A priori* metastable atoms experience a van der Waals interaction from both edges of the slit. Because of the relatively large width of the slit the two edges act independently leading to symmetric deflections of the inelastically scattered atoms. In addition, as the $^3P_2 \rightarrow^3 P_0$ channel is energetically closed, the only contribution to the signal originates from the incoming $^3P_0$ population. Time of flight spectra (Fig. 1) have been accumulated for a series of scattering angles ranging from 40^o up to 75^o, at several selected incident velocities : 535m/s and 356m/s for argon, 436m/s for krypton. In order to prevent any experimental drift during the long acquisition times (about 7 hours for each angle) this series of spectra has been calibrated in relative values by measuring the intense uv-photon peak present at all angles over a shorter time (1000-1500s). By this procedure the $^3P_0 - ^3P_2$ differential cross sections have been determined in relative values. They are shown in Fig. 2 . The small shift (about $+2^o$) of their maximum with respect to the theoretical deflection angle can come from a slight misalignment of the normal to the slit with respect to the beam axis or from the fact that the slit edge is not exactly perpendicular to the slit plane. Another possible explanation is the corrugation of the metallic wall along the direction $\hat{x}$ parallel to the beam axis: only those parts of the wall the slope of which is positive can be approached by the atoms, which gives a systematic positive shift to the deflection angle.

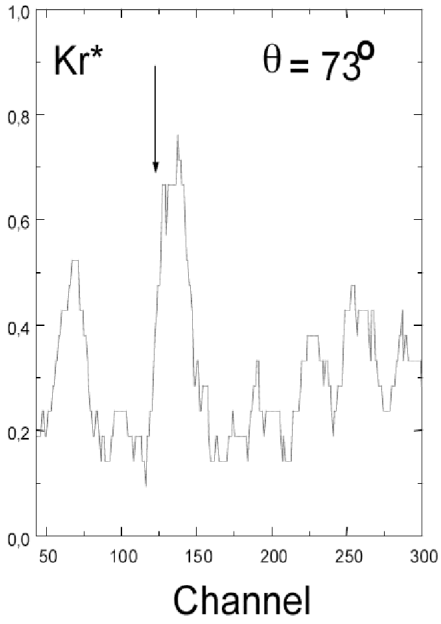

Fig. 1. Time of flight spectrum of Kr* atoms at $\theta_f = 73^o$. Vertical arrow: predicted inelastic tof.

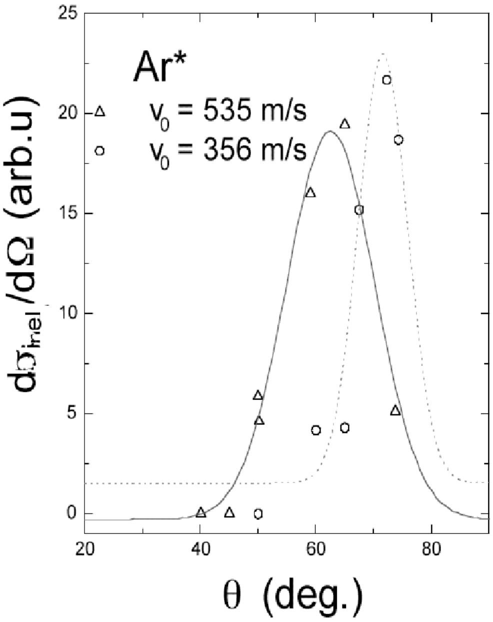

Fig. 2. Angular distribution of inelastically scattered Ar* atoms: $v_0 = 535$m/s (full line), 356m/s (dotted line).

[1] M. Ducloy, in *Nanoscale Science and Technology*, (Kluwer, The Netherlands, 1998), p. 235.