

## Brief Reports

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Collisional depopulation of rubidium Rydberg states by ND<sub>3</sub>

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This paper reports the measurement of the depopulation cross sections of the Rb(*ns*) and Rb(*nd*) states by ND<sub>3</sub> ( $23 \leq n \leq 50$ ). The experimental data are discussed with reference to previous work done on collisions between Rb(*ns*) and Rb(*nd*) states and NH<sub>3</sub>.

Several experimental investigations have demonstrated the importance of near-resonant energy transfer in the collisional depopulation of high Rydberg states by polar molecules.<sup>1-4</sup> For example, the quenching cross sections of highly excited rubidium atoms by CO have been measured and compared to the predictions given by the impulse approximation (IA).<sup>3</sup> The good agreement observed between experimental and theoretical values has already shown the importance of the internal structure of the molecular target

and of the long-range Rydberg-electron-dipole interaction.

Collisions of rubidium Rydberg states with ammonia<sup>4</sup> have been recently studied, and the following observations were made. (a) The NH<sub>3</sub> inversion can produce a very efficient *l*-mixing process. For instance, when the energy difference between the *n* initial state and the neighboring manifold matches the NH<sub>3</sub> inversion energy  $\sim 0.7 \text{ cm}^{-1}$  [case of the Rb(48*d*) state], the depopulation cross section is about five times larger than the depopulation cross section measured when this resonance condition is not fulfilled [case of the Rb(*nd*) levels,  $n < 40$ ]. (b) The IA provides the correct *n* behavior of the depopulation cross section.

Here we report the extension of our previous measurements of the collisional depopulation of the Rb(*ns*) and Rb(*nd*) states with NH<sub>3</sub> to include the deuterated ammonia ND<sub>3</sub>. We wish to confirm our description of the role played by the NH<sub>3</sub> inversion. As the ND<sub>3</sub> inversion energy<sup>5</sup> is now  $0.052 \text{ cm}^{-1}$  one can predict, on the basis of our previous work, that the *l*-mixing process of Rb(*ns*) and Rb(*nd*)

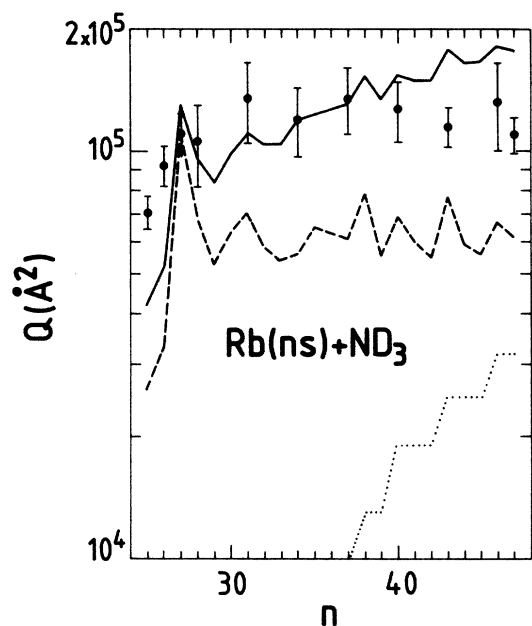


FIG. 1. Depopulation cross sections of the Rb(*ns*) states in collisions with ND<sub>3</sub>. Experimental data (for *ns* and *nd* levels) have been normalized against IA calculations for the Rb(34*s*) state. The collisional ionization appears as a dotted line. The dashed line represents the sum of the *n* changing and ionization processes. The total depopulation (including *l* mixing, *n* changing, and ionization) is shown as a solid line. Crude estimations of the absolute data are in rough agreement with IA calculations.

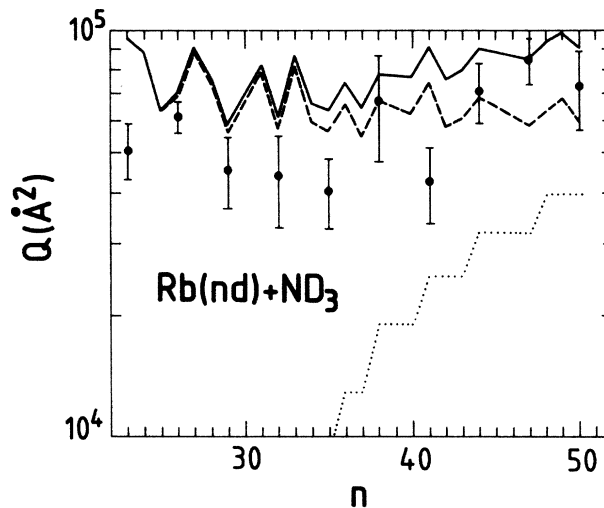


FIG. 2. Depopulation cross section of the Rb(*nd*) states in collisions with ND<sub>3</sub> as in Fig. 1.

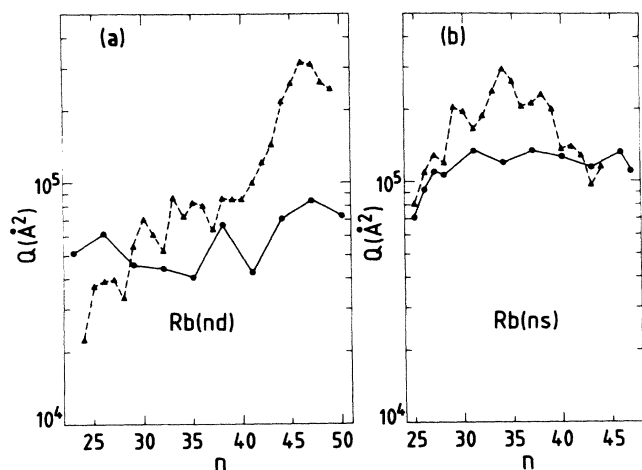


FIG. 3. Experimental quenching cross sections for Rb(*nd*) and Rb(*ns*) states [(a) and (b), respectively] colliding with NH<sub>3</sub> (triangles) or ND<sub>3</sub> (circles). Results for NH<sub>3</sub> are from Ref. 4. The dashed and continuous lines between the points are just drawn for clarity. The error bars are not reported (see Figs. 1 and 2 for ND<sub>3</sub> or Ref. 4 for NH<sub>3</sub>).

states ( $n \sim 23-50$ ) induced by the ND<sub>3</sub> inversion cannot affect appreciably the total depopulation, since its energy balance remains too important ( $\geq 0.7 \text{ cm}^{-1}$ ). The depopulation cross section is then expected to be approximately constant and large ( $\sim 10^5 \text{ Å}^2$ ), since the ND<sub>3</sub> dipole moment is approximately equal to that of NH<sub>3</sub>.<sup>6</sup>

The experimental method is described in detail else-

where.<sup>4,7</sup> It involves the measurement of the effective lifetime of the Rb(*ns*) or Rb(*nd*) state as a function of ND<sub>3</sub> density. This is achieved by a time-resolved selective field ionization technique. As was the case for NH<sub>3</sub> absolute pressure measurements appear hard to perform accurately.<sup>7</sup> Our experimental procedure allows only the study of the relative  $n$  behavior of the cross section. Finally, a crude estimation of the absolute pressure obtained by the calibration of the ionization gauge (located after the cell) against the capacitance manometer seems to indicate that the obtained results are in rough agreement (within a factor of 3) with IA calculations.

In Figs. 1 and 2 are plotted the cross sections versus  $n$  for the *s* and *d* states, respectively. The experimental data (for *ns* and *nd* levels) have been normalized against the IA calculations for the Rb(34*s*) state.<sup>8</sup> Figures 1 and 2 show that the quenching cross section remains approximately constant as predicted by the IA. This clearly supports our previous description of the  $n$  behavior of the Rb Rydberg-state depopulation cross sections by NH<sub>3</sub>.

Finally, Fig. 3 shows a comparison between the experimental results obtained with NH<sub>3</sub> and ND<sub>3</sub>. The  $n$  behavior of both sets of data clearly exhibits the importance of NH<sub>3</sub> inversion on the depopulation cross section. For ND<sub>3</sub>, as previously mentioned, no resonance occurs in the investigated range of  $n$  values. The isotope effect appears more pronounced for Rb(*nd*) states than for Rb(*ns*) states. This is perhaps due to the fact that the *ns* levels have a more penetrating character. In this case the validity of the IA approach taking into account only the  $e^-$ -molecule interaction is probably more questionable. This point clearly calls for further theoretical studies.

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<sup>2</sup>See also recent reviews by M. Matsuzawa and by F. B. Dunning and R. F. Stebbings, in *Rydberg States of Atoms and Molecules*, edited by R. F. Stebbings and F. B. Dunning (Cambridge Univ. Press, Cambridge, 1983).

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