# Experimental Differential Cross Sections for Laser-Excited Sodium in the $^2P_{3/2}$ State in Collision with Mercury

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Differential cross sections for Na $^2P_{3/2}$ have been measured in collisions with Hg at thermal energies. In the angular range between 4° and 30°, the coarse structure of the cross section relative to the well-known ground-state interaction is given. In the region between 40° and 70°, an oscillatory behavior of the cross section is observed. Because of their velocity dependence, these oscillations are attributed to fine-structure transitions.

In this Letter we report an experiment with a molecular beam of Na in the $^2P_{3/2}$ state in collisions with Hg. In this experiment the differential cross sections have been measured at thermal energies with a high resolution ($\Delta v/v = 2.5\%$). The excitation has been generated with a laser. From such experiments it should eventually be possible to determine the intermolecular potential for the excited state with the same degree of accuracy as for the ground state. At the present time the most reliable results with the same goal seem to stem from fluorescence-cell experiments.[1-3] In the evaluation of these experiments one considers the behavior of integral cross sections averaged over a broad distribution of relative velocities. Molecular-beam experiments should improve the accuracy of the evaluation, especially if differential cross sections with a narrow distribution of velocities are available for analysis.

For ground-state particles, similar measurements have been evaluated previously to yield potentials with an accuracy of a few percent over a wide range of internuclear distances.[4] When extending these experiments to the excited states, the conventional molecular-beam technique has to be refined (1) in the production of the beam of the excited species and (2) in the careful design and consideration of the resolution of the experiment.

The excitation of the Na beam has been achieved with a cw-dye laser tuned to the frequency for the transition between the $F = 2$ level in the ground state and the $F = 3$ level in the $^2P_{3/2}$ state.[5,6] The efficiency of this excitation has been determined by a measurement on the deflection of the molecular beam due to the momentum of the photons.[5] Since in our experiment the beam is well collimated and selected with respect to the velocity, this momentum transfer is clearly visible as a splitting of the original single-peak beam profile (with the laser turned off) into one with two distinct peaks (with the laser turned on). Details of this experiment will be given elsewhere.[7] For our purpose it is sufficient to mention that the analysis of these experiments at various velocities shows that, on the average, 55% of the beam particles underwent excitation processes. This is close to the possible upper limit of 62.5% associated with the population of the $F = 2$ level in the Na beam. This result is obtained with a laser power of 30 mW, which is enough to reach saturation. This means that induced emissions occur more frequently than spontaneous decays. Based on the semiclassical theory, one finds a period of about $5\times10^{-10}$ sec for the excitation cycle. In principle, the incoming channel is a superposition of the ground and excited states.[8] But since the total wave functions for the excited and ground states differ in their nuclear part and the bandwidth of the laser (30 MHz) is such that the Na is out of resonance for internuclear distances less than a certain (large) radius $R_c$ (because of the distortion by the approaching target), we neglect the cross terms in this superposition. From calculations of the potentials,[9] we estimate the $R_c$ for Na-Hg to be at least 50 a.u. Consequently, only half of the atoms undergoing excitation are considered to be in the excited stated during the collision. This holds independently of the excitation mechanism. For our experiment, we can expect approximately 27.5% of the beam interacting under the potential of the excited state.

Since the scattering volume for the excited state and for the ground state will be different from each other in general, the determination of the cross sections from the experimental data poses a serious problem. Denoting by $F(g,\theta)$ the distribution which applies for the ground-state interaction,[10] by $F^*(g,\theta)$ the distribution for the interaction in the excited state, by $\sigma_x$ the ground-

state cross section, and by $\sigma^*$ the excited-state cross section, one can write the experimentally measured quantities as follows:

$$I_{OL} = \int F(g,\theta)\sigma_x dg\, d\theta, \tag{1}$$

$$I_{ML} = \int F(g,\theta)\sigma_x dg\, d\theta + \int F^*(g,\theta)(\sigma^* - \sigma_x) dg\, d\theta, \tag{2}$$

$$\Delta = \int F^*(g,\theta)(\sigma_x - \sigma^*) dg\, d\theta, \tag{3}$$

where $I_{OL}$ denotes the intensity without the laser; $I_{ML}$, the intensity with the laser; and $\Delta$, their difference. We integrate over the relative velocity $g$ and the center-of-mass angle $\theta$. The widths of the distributions $F$ and $F^*$ depend on various parameters, but our Monte Carlo calculations indicate that for our experiment the spatial distributions along the primary beam are dominant. Then the distribution $F$ is determined by the distribution of Hg; and $F^*$, by the product of the Hg distribution with the intensity profile of the laser.

In the signal with the laser [Eq. (2)], we see three terms contributing: the ground-state cross section $\sigma_x$ multiplied by the original distribution $F$, $\sigma_x$ multiplied by the distribution of the excited atoms $F^*$, and finally the excited-state cross section $\sigma^*$ multiplied by $F^*$. In the difference signal we find both cross sections averaged with the distribution $F^*$. Therefore, for the determination of the exact cross section of the excited state, one must know the ground-state cross section averaged over the distribution of excited particles. For a quantitative evaluation, this distribution must be known with a higher accuracy than the difference signal, which is on the order of 10% of the ground-state signal. But for the purpose of the evaluation, specific structures in the cross section which can be determined without the exact knowledge of the distribution functions are usually sufficient. These can be found in the difference signal if one takes into account the general behavior of the cross sections. If one of the terms in the difference signal shows strong oscillations in a certain angular range and the other term is monotonic, the difference signal will yield these oscillations unambiguously [see Eq. (3)].

A second possibility to determine parts of the cross section arises if the difference signal has zeros. This occurs where the cross section $\sigma_x$ is equal to $\sigma^*$. If, in addition, the distribution functions $F$ and $F^*$ are approximately equal, one can give for each zero a specific point where the ground-state and excited-state cross sections, averaged over $F$, are the same.

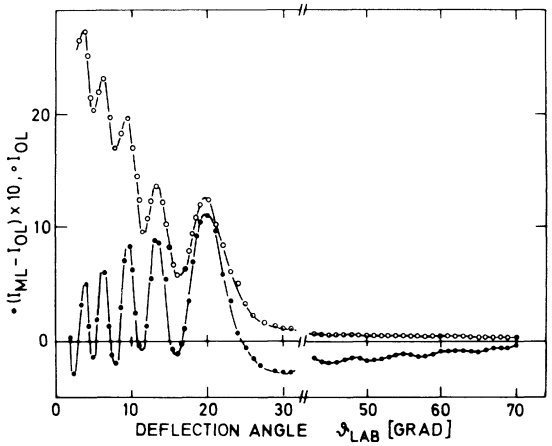


FIG. 1. Differential cross sections for the ground-state interaction (○) and the difference signal (●).

A direct evaluation of the difference signal in comparison with the calculations has also been suggested.[11] But it is obvious that the data remain extremely sensitive to the experimental averaging process. These considerations have to be kept in mind in discussions of the experimental results. In Fig. 1, we show the signal from the ground state [Eq. (1)] together with the difference signal [Eq. (3)] between 4° and 70° for a barycentric energy of $43.5 \times 10^{-14}$ erg. Two sections are displayed: a small-angle part from 4° to 30° and a large-angle part from 40° to 70° (laboratory angles). The small-angle part is characterized by the rainbow structure for the ground-state interaction which is well understood in terms of the respective interaction potential.[4] In the difference signal one finds oscillations of a large amplitude which are exactly in phase with the rainbow oscillations. From this we conclude that these oscillations are due to the ground-state contribution to the difference signal [Eq. (3)] and that a possible contribution from the excited state is either monotonic or has a relatively small amplitude. This conclusion is in contrast to a report on a similar experiment from another laboratory on the Na-Ne interaction.[11] For this reason, we can deduce from the measured signal only an approximate cross section, using the condition that in the zeros of the differential signal, the cross sections of the excited and the ground state are equal. With approximate corrections for the small difference in the distribution functions, this condition yields the results displayed in Fig. 2, where the differential cross sections of the excited state are given in relation to those

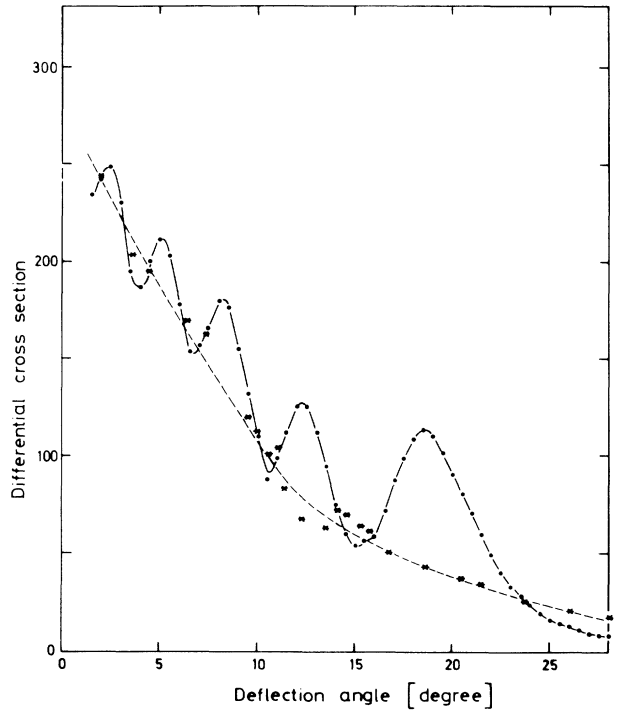


FIG. 2. Differential cross sections for the ground-state interaction (solid line) and coarse structure of the cross sections for the $^2P_{3/2}$ state (dashed line).

for the ground state. As discussed above, the broken line connecting the points from this evaluation gives only the coarse structure of the cross section for the excited state.

In contrast to this, the structure of the excited-state cross section in the large-angle part can be given precisely, because the ground-state cross section is monotonic there and the structure observed can be unambiguously attributed to the excited state. The contribution is clearly seen in Fig. 1; and for another energy, after subtracting a fraction of the ground-state signal we have the result shown in Fig. 3.

The attempt to interpret these data with the differential cross sections for calculated potentials[9] in terms of a completely elastic calculation fails. Specifically, the spacing of the rainbow oscillations of the calculation turns out to be too large by a factor of 2, which is outside the errors of the measurement. The agreement improves if parts of the inelastic effects for transitions between the $^2P_{3/2}$ and the $^2P_{1/2}$ states are taken into account. These findings agree well with a recent calculation by Reid[12] for Na-He, where the influence of such transitions has been investigated, taking the couplings properly into account. In

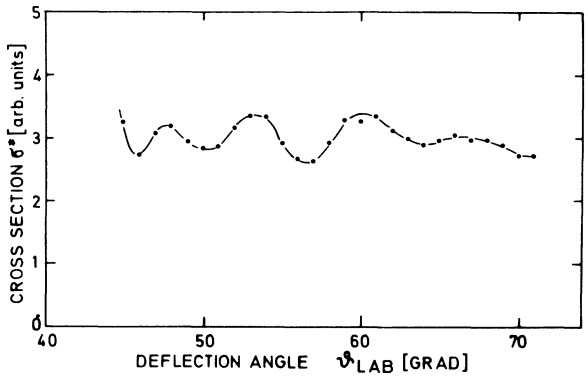


FIG. 3. Differential cross sections for the $^2P_{3/2}$ state.

these results the transitions are shown to cause oscillations similar to the ones observed here. An extrapolation from the Na-He calculation to the Na-Hg experiment is of questionable validity; however, a full calculation is presently being performed in our laboratory.

Another experimental result extracted from our data supports the importance of inelastic effects. The positions of the extrema in the large-angle range for various energies can be reduced by the relation $\theta g = \text{const}$. This is typical of the inelastic processes involved in fine-structure transitions. In the elastic case, one would expect $\theta E = \text{const}$ to hold and the observed pattern to be due to rainbow scattering.

In summary, we have established some parts of the differential cross section for $^2P_{3/2}$ Na in collision with Hg. The coarse structure can be given for the low-angle part where the influence of the experimental distribution is found to obscure the details of the cross section. In the large-angle part where this influence can be removed, we have found oscillations to determine the cross section of the excited state. By comparison with the calculated cross sections and by the $1/g$ dependence of these oscillations we are led to the conclusion that these are due to fine-structure transitions. To our knowledge, this has been observed for the first time in the differential cross sections in a molecular-beam experiment.

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# Quasilinear Theory of Ion-Cyclotron-Resonance Heating of Plasmas and Associated Longitudinal Cooling*

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It is shown from a quasilinear theory that an initially isotropic magnetized plasma will be forced into an anisotropic state in ion-cyclotron-resonance heating. Strong heating of the perpendicular ion temperature and strong cooling of the longitudinal temperature should occur simultaneously. The maximum temperature ratio predicted by the quasilinear theory is in exact agreement with that predicted from the basic thermodynamic arguments by Busnardo-Neto, Dawson, Kamimura, and Lin. Heating by fast hydromagnetic waves is also examined.

Very recently Busnardo-Neto, Dawson, Kamimura, and Lin[1] have reported on some studies of the absorption of ion-cyclotron waves which involve computer simulations and a theoretical analysis. As opposed to previous reports, they find that an initially isotropic plasma is forced into an anisotropic state by means of an applied external pump field near the ion-cyclotron frequency. Their studies showed that in addition to a strong heating of the ions in the perpendicular direction (perpendicular to the external magnetic field $\vec{B} = B\hat{i}_z$) there was a strong cooling of the parallel ion temperature. They used very basic thermodynamic arguments to predict the nonlinear saturation of the heating-cooling process; and the simulation results confirmed their predictions. Their argument runs as follows: An ion-cyclotron wave of frequency $\omega$ and wave number $k$ propagating along an external magnetic field resonates with particles of velocity $v_R$ such that $kv_R = \omega - \omega_C$, where $\omega_C$ is the ion-cyclotron frequency. Since the frequency of these waves is always less than the ion-cyclotron frequency, the resonant particles and the wave propagate in opposite directions. The resonant particles either continually gain or lose energy to the wave. If we assume that the particle absorbs $n$ quanta from the wave, its perpendicular energy will increase by $n\hbar\omega_C$. The wave supplies only $n\hbar\omega$ of this energy; thus the remainder $n\hbar(\omega_C - \omega)$ must come from the parallel motion of the particle. In other words in absorbing energy $dW$ from the wave the perpendicular energy increases by $dQ_\perp = dW\omega_C/\omega$, whereas the parallel energy decreases by $dQ_\parallel = dW(\omega_C - \omega)/\omega$. From the thermodynamic point of view, one can regard the parallel and perpendicular motions as two independent systems with two temperatures $T_\parallel$ and $T_\perp$. Since the entropy $S$ must increase or at best remain the same for a reversible process, one finds that $dS = -dQ_\parallel/T_\parallel + dQ_\perp/T_\perp \geq 0$. That is, $\omega_C/(\omega_C - \omega) \geq T_\perp/T_\parallel$. This is an upper limit on the temperature anisotropy that can be achieved. It is my aim in this paper to give a rigorous proof of these results from the classical quasilinear plasma kinetic theory (CQPKT).[2-4]

According to the quasilinear theory [for example, see Eq. (52) of Ref. 2 and Eq. (11-33) of Ref. 3], one can show that the time rate of change of the particle distribution function due to emission and absorption of cyclotron waves of frequency $\omega$ and wave vector $\vec{k}$ in a box of volume $L^3$ is given