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Polarization Effects in the Differential Quenching Cross Section of Na* by Diatomic Molecules

I. V. Hertel, H. Hofmann, and K. A. Rost
Fachbereich Physik der Universität Kaiserslautern, 6750 Kaiserslautern, West Germany
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The dependence of scattering intensities for the quenching process $Na^*(3^2P) + N_2(v = 0) \rightarrow Na(3^2S) + N_2(v')$ on the polarization of the exciting laser light has been studied. A distinct anisotropy is observed at small scattering angles, which is most pronounced where the scattering cross section is largest. It is found that the quenching cross section σ_1 for a $3p\pi$ state is smaller that σ_0 for the $3p\sigma$ state and thus electronic angular momentum is transferred to collisional and/or rotational angular momentum.

In a recent Letter[1] we have reported first experiments on the quenching of laser-excited $Na(3^2P_{3/2})$ at thermal energies in the differential scattering process:

$$Na(3^2P_{3/2}) + N_2(v=0, j_{therm}) + E_{therm} \rightarrow Na(3^2S_{1/2}) + N_2(v', j') + E_{kin}'. \quad (1)$$

A crossed-beam apparatus is used. The primary sodium beam is supersonic with a full width at half-maximum (FWHM) of 20%, and the thermal molecular beam effuses from a capillary array at 80 or 300°K. The initial c.m.-system energy, E_{therm}, is defined by the relative average energies and kinematics of the two beams. The amount of energy transferred into vibrational and/or rotational energy is found from the measured final kinetic energy E_{kin}' and from the electronic excitation energy E_{el} (= 2.1 eV), and is $\Delta E_{vib\ rot} = E_{el} + E_{therm} - E_{kin}'$. A typical energy-transfer spectrum is shown in Fig. 1, displaying the c.m.-system kinetic energy after collision.

It has been stated that the experimental findings are in qualitative agreement with theoretical models proposed by Bjerre and Nikitin[2] and by Bauer, Fisher, and Gilmore.[3] The theoretical calculations are tentative for several reasons. Among other shortcomings, no attention has been paid to the influence of rotational-energy transfer, and the general assumption is that only vibrational energy levels are excited (the v' scale in Fig. 1 corresponds to this assumption). The present

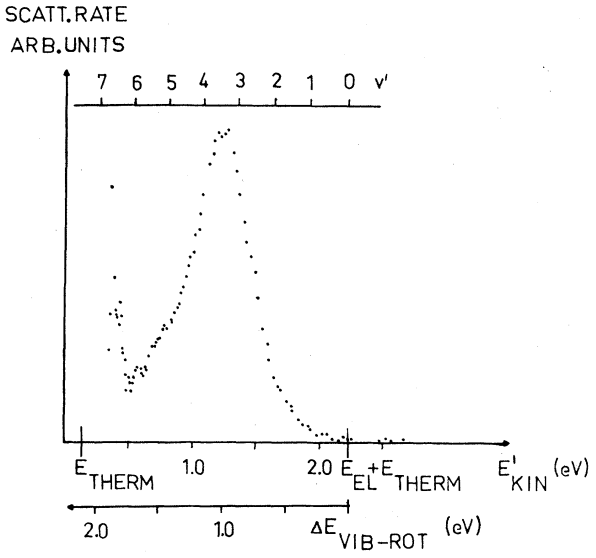

FIG. 1. Energy-transfer spectrum for the quenching process $N_2 + Na^*(3P) \rightarrow N_2 + Na(3S)$. Displayed is the difference "light on" – "light off" as a function of the energy after collision in the c.m. system. Initial Na velocity is 1370 m/sec, 20% FWHM; N_2 temperature is ≈ 80° K, $\theta_{lab} = 10°$ corresponding to $\theta_{c.m.s.} = 5°$ at $\Delta E_{vibrot} = 1$ eV. The rise of the scattering rate below 0.5 eV indicates elastic scattering from the excited sodium which cannot be distinguished unambiguously from a remaining energy transfer to $v' = 7$.

experiment does not allow us to distinguish between transitions among the vibrational and/or rotational levels, as long as they belong to the same $\Delta E_{\text{vib rot}}$. However, a numerical study of the kinematics and resolution of the present experiments indicates that pure transitions with $\Delta j = 0$, $\Delta v \neq 0$ would lead to observable structures on positions indicated by v'. The absence of these distinct peaks illustrates the need for a less simplified model.

Another aspect, hitherto undiscussed, is the possible influence of orientation and alignment of the excited sodium atom. With respect to the c.m. system, the $3p$ electron may be either in the $3p\sigma$ or in the $3p\pi$ state. Consequently, the quenching cross section σ_0 for the $3p\sigma \rightarrow 3s\sigma$ transition (with $\Delta M = 0$) has to be distinguished from σ_1 for the $3p\pi \rightarrow 3s\sigma$ quenching (with $\Delta M = \pm 1$). The present experiment is a first attempt to attack both these problems indicated, which, as it turns out, are closely related.

Electron scattering experiments by laser-excited Na atoms[4,5] have demonstrated that the study of scattering intensities as a function of the direction of the exciting-laser-light polarization is a powerful tool to reveal details of the scattering dynamics. Very large anisotropies in the differential electron scattering by Na* are observed for $3p \rightarrow ns$ transitions and can be evaluated in terms of scattering amplitudes for $\Delta M = 0, \pm 1$ transitions. No such effects have been reported in the elastic differential heavy-particle scattering of Na(3^2P) by Ne[6] and by Hg.[7] While this may partially be attributed to the large variety of possible transitions between the magnetic sublevels in a $3p \rightarrow 3p$ transition, one might expect some measurable effects for the process under discussion, which is a $3p \rightarrow 3s$ transition.

By optical pumping with linearly polarized light the atom is prepared in an aligned state with even multipole moments $w(0), w(2), \ldots$.[8] The ratio $w(2)/w(0)$, which is proportional to the measurable fluorescence intensity of spontaneous decay, can vary between the values 1.47 and 1.74.[9] The measured value of 1.47 thus illustrates the influence of incomplete optical pumping and radiation trapping.

We now summarize the experimental observations of polarization effects in the differential $\text{Na}(3^2P_{3/2}) + N_2$ quenching process.

(i) A definite but small anisotropy of the differential quenching cross section is observed, when the electric vector $\vec{E}$ of the exciting laser light is rotated in the scattering plane. The measured scattering intensity may be written as

$$I^{sc} \propto 1 + \alpha - \beta \cos 2\psi - \gamma \sin 2\psi, \quad (2)$$

where ψ is the angle of E with respect to the c.m. system before the collision [from Ref. 9, Eqs. (17), (49), (50)]. Three typical examples are shown in Fig. 2.

(ii) The anisotropy as a function of the energy transfer seems to follow the energy-transfer spectrum (Fig. 1) as shown in Fig. 3, where

$$\frac{I_{\max}{}^{sc}}{I_{\min}{}^{sc}} = \frac{1 + \alpha + (\beta^2 + \gamma^2)^{1/2}}{1 + \alpha - (\beta^2 + \gamma^2)^{1/2}}$$

is plotted together with 1-standard-deviation error as obtained from least-squares fits to the measured curves [Eq. (2), Fig. 2]. At the maximum the effect is ~18% for quenching by N_2 at a temperature of $T \approx 80$°K. The anisotropy vanishes in the wings of the energy-transfer spectrum.

(iii) The anisotropy seems not to depend significantly on the scattering angle in the angular

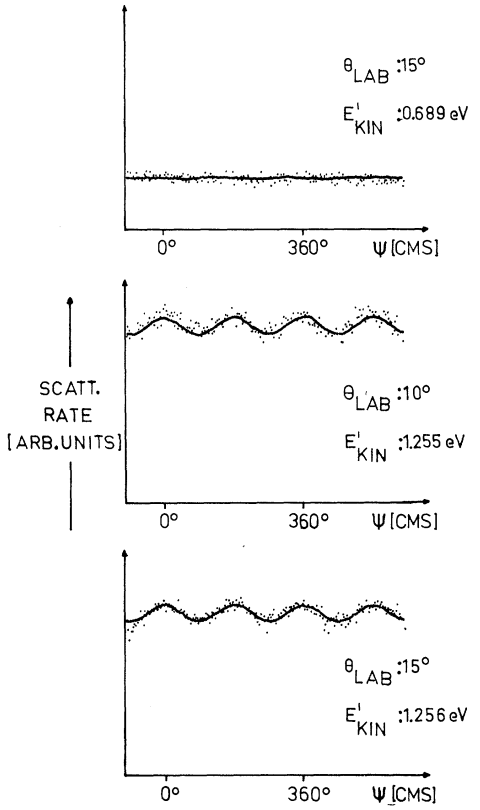

FIG. 2. Scattering intensity for $Na^* + N_2$ quenching as a function of the polarization angle with respect to the c.m. system for three different θ_{lab}, ΔE_{vibrot}, together with a least-squares fit according to Eq. (3).

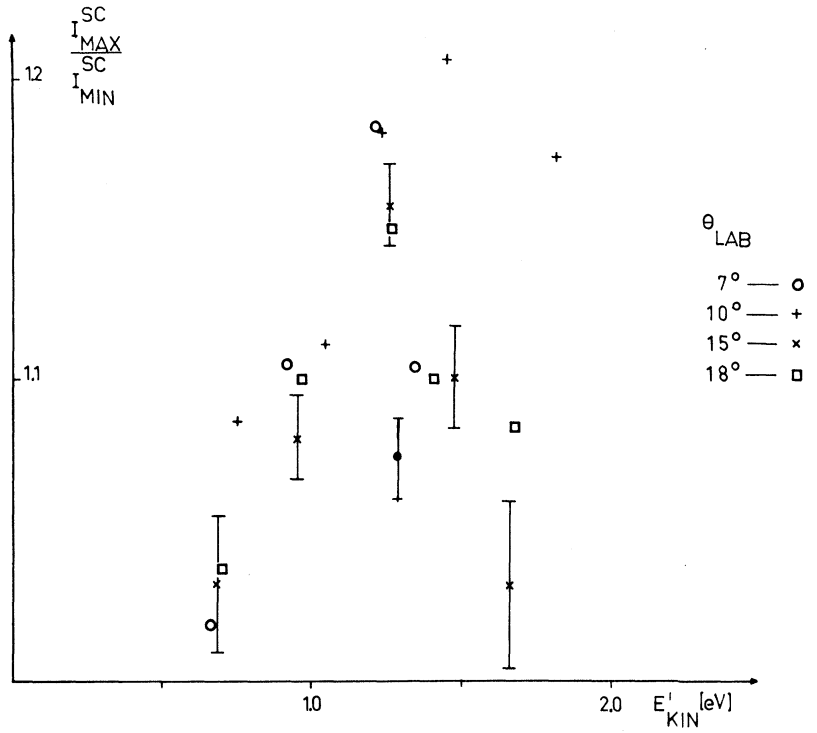

FIG. 3. Ratio of maximum to minimum scattering intensity taken from fits (as shown in Fig. 2) as a function of the energy transferred to N_2. The laboratory scattering angles given correspond at $E_{vibrot} = 1$ eV to $\theta_{c.m.s.} = 1.3°$ (○), 5° (+), 11° (×), and 15° (□) for 80°K and to $\theta_{c.m.s.} = 1.8$ (●) at 300°K.

range under investigation.

(iv) The quenching cross section has its maximum when the $\vec{E}$ vector is approximately parallel to the c.m. system, i.e., when $\gamma = 0$ and $\beta < 0$. This seems to change somewhat in the wings of the energy-transfer spectrum. There, however, the experimental errors in determining γ/β are largest.

(v) Measurements on a circular anisotropy, i.e., a change of the cross section for σ^+ and σ^- light excitation at $\Delta E_{vib\ rot} = 1$ eV, $\theta_{lab} = 18°$, $\theta_{c.m.s.} = 14.7°$, have shown no significant effect ($\leq 4\%$).

(vi) By a change in the temperature of the molecular beam, its distribution of rotational states is altered significantly, while E_{therm} is mainly determined by the sodium velocity. At 300°K the linear anisotropy decreases to about half the value at 80°K. One typical point is shown in Fig. 3 (solid circle with error bar).

(vii) Similar observations have been made for the quenching of Na* by D_2. For O_2, CO, and CO_2, measurements have only been carried out for 300°K where no anisotropy has been observed.

At present, we do not attempt to evaluate the measured polarization anisotropy quantitatively in terms of scattering amplitudes or scattering multipole moments. Several experimental uncertainties prevent us from doing so: (a) The value of the atomic alignment averaged over the collision region is not known. The fluorescence anisotropy gives only an approximate value, since the light detector does not see the total scattering volume. The value, however, has been used to apply a first-order correction to the experimental data, thus given in Fig. 3. (b) The c.m. system is defined only to $\pm 6°$ since the N_2 beam is effusing from a capillary array and has a modified Maxwellian velocity distribution. Model computations, however, show that this does not influence the polarization measurements as long as γ/β does not change too quickly with the collision angle. Under these conditions the N_2 beam is represented adequately by its most probable velocity. (c) For a full analysis, measurements should have been performed also with the laser incident in the scattering plane, which is experimentally somewhat difficult to realize. Therefore we restrict ourselves to a qualitative comparison to the electron-scattering case and will give a more refined analysis in connection with additional experimental data in a forthcoming

publication.

As in the electron-scattering case the quenching cross section σ_0 of a $3p\sigma$ state (cigar-shaped) is larger than the cross section σ_1 of a $3p\pi$ state (disk-shaped) in the angular range accessible. The values of anisotropy are much smaller, however. The $3p\pi$ quenching necessitates the destruction of orbital angular momentum, the final atomic state being $3s\sigma$. These findings indicate that in the heavy-particle-collision experiment both disk- and cigar-shaped charge distributions of the atom exhibit cross sections of less pronounced difference than in the electron-scattering case.

Since the total-angular-momentum projection with respect to the c.m. system as a quantization axis has to be conserved, the electronic angular momentum must be converted into either relative angular momentum (L_{coll}, M_{coll}) of the collision or into rotational angular momentum of the molecule.

In general the scattering amplitudes describing the process $3p\pi \rightarrow 3s\sigma$ have to include the rotational quantum numbers j, m_j, j', and m_j' before and after the collision. Conservation of the total angular momentum enforces $M+m_j=m_j'+M_{\mathrm{coll}}$. While in the e-Na* case only a few partial waves participate in the scattering process, and for a change in M finite collision angles are necessary, in the quenching by heavy particles a large number of partial waves contribute at small scattering angles. In addition, transitions in the rotational projection quantum numbers can occur. Whether angular momentum actually is transferred into rotation is an open question. The strong temperature dependence of the anisotropy clearly indicates the influence of rotational m_j changes (at $T=80$°K the most probable j is ~3, while for 300°K it is around 7). In summation over a wide range of j, m_j, and M, the resulting cross section for a $3p\pi \rightarrow 3s\sigma$ transition may lead to a less pronounced anisotropy compared to the electron-scattering case.

Let us finally try to hint at an explanation of the fact that the anisotropy is largest where the quenching cross section has its maximum: In an involved curve-crossing process, the largest cross section indicates the most direct process, and thus the initial alignment of the atom influences the cross section most strongly. Where the process itself is less probable the memory is lost during the collision time and $3p\pi$ and $3p\sigma$ quenching may become equally probable. An enhanced rotational momentum transfer provides for the conservation of angular momentum.

We hope to have illustrated some new aspects of the process of energy transfer from electronic to rotational-vibrational-translational in atom-molecule collisions and to stimulate new theoretical investigations. Such studies would have to include rotational energy transfer and should explain how electronic angular momentum is transferred into rotations and/or collisional angular momentum.

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