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Collisional Excitation Of N+ At 50 KeV

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in a two-level system for SF_6.

The pulse-delay times shown in Fig. 3 refer to the intensity peaks; they cannot be accounted for theoretically, even if we were dealing with SIT in a two-level system, since the dephasing collision times and laser-pulse durations were of the same order of magnitude in our studies. We find a decrease in delay time with decreasing chlorine pressure at constant total values of p_{Cl_2} corresponding to a decrease in pulse-delay time with increasing dephasing collision frequency (see Fig. 3).

We did not observe multiple-pulse formation and nearly complete transparency, perhaps because of relatively poor laser-beam quality. Gibbs and Slusher[7] showed in studies of SIT for a two-level system that, for a single-mode laser output, the intensity distribution must be uniform in order to allow observation of multiple pulses and peak amplification.

In conclusion, we re-emphasize the fact that we are dealing with a "pseudo-two-level" system since the upper state in the bound-free transitions is unstable in times of the order of 10^{-12} sec. The fact that anomalous transmission is nevertheless observed must indicate that nonlinear interactions with the coherent radiation field take place on a similar time scale, thereby producing excessive transparency in a kind of lossy system. The detailed nature of this problem clearly requires theoretical study.

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*Much more extensive compilations of experimental findings and of related interpretive work appear in the Ph.D. thesis of R. C. Sepucha, University of California, San Diego, 1971.

[1]S. L. McCall and E. L. Hahn, Phys. Rev. Lett. 18, 908 (1967).

[2]S. L. McCall and E. L. Hahn, Phys. Rev. 183, 457 (1969).

[3]C. K. Rhodes, A. Szöke, and A. Javan, Phys. Rev. Lett. 21, 1151 (1968).

[4]L. E. Estes, D. C. Eteson, and L. M. Narducci, IEEE J. Quantum Electron. 6, 546 (1970).

[5]F. A. Hopf and M. O. Scully, Phys. Rev. 179, 399 (1969).

[6]C. K. N. Patel and R. E. Slusher, Phys. Rev. Lett. 19, 1019 (1967).

[7]H. M. Gibbs and R. E. Slusher, Phys. Rev. Lett. 24, 638 (1970).

[8]G. E. Gibson and N. S. Bayliss, Phys. Rev. 44, 188 (1933); G. E. Gibson, O. K. Rice, and N. S. Bayliss, Phys. Rev. 44, 143 (1933); P. Sulzer and K. Wieland, Helv. Phys. Acta 25, 653 (1952); R. L. Wilkins, J. Quant. Spectrosc. Radiat. Transfer 4, 775 (1964); T. A. Jacobs and R. R. Giedt, J. Quant. Spectrosc. Radiat. Transfer 5, 457 (1965); S. S. Penner and D. B. Olfe, *Radiation and Reentry* (Academic, New York, 1968), Chap. 1.

Collisional Excitation of N^+ at 50 keV†

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The excitation spectrum of N^+ has been observed by examining the energy lost by a 50-keV N^+ beam passing through a He target. The spectrum exhibits dramatic features with large cross sections. Determination of the approximate ratio of metastable to ground-state ions in the primary ion beam has permitted measurement of excitation cross sections from both ground-state and metastable N^+ ions colliding with He target atoms.

Energy analysis of fast ions after collision has recently been employed to study excitations of atoms, molecules, and ions.[1-3] Park and Schowengerdt[4] describe the apparatus employed in the present study. Modifications have improved energy resolution and facilitated data handling,[5] but have not altered the basic technique.

The ion N^+ is of interest in astrophysics and atmospheric physics. The forbidden transition $2p^2\,{}^1D \rightarrow 2p^2\,{}^3P$ at 6584 Å is used extensively for determination of electron temperature and degree of ionization in nebulas.[6-8] This transition and the forbidden transition $2p^2\,{}^1S \rightarrow 2p^2\,{}^1D$ at 5755 Å are also observed in aurora and airglow and are employed in understanding these phenomena.[9,10]

Beam-foil spectroscopy measurements of lifetimes of excited N^+ states,[11] electron impact ionization of N^+,[12] and emission lines of N^+ from electron impact on N_2,[13] have been reported.

The apparent differential (energy-loss) cross

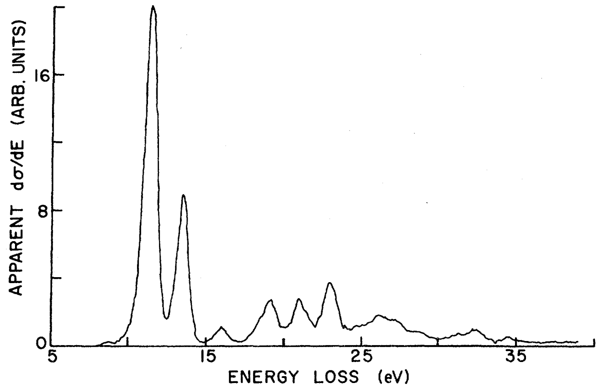

FIG. 1. A typical data set for N^+ on He at 50 keV, showing the apparent cross-section differential in energy loss versus the energy lost by the N^+ ion. The term apparent means that the instrumental resolution function has not been unfolded from the data. The data have been normalized with respect to the magnitude of the incident current and the target particle density.

section for 50-keV N^+ incident on He is shown in Fig. 1. Helium was chosen as a target because the lowest-lying excitation of helium, requiring 19.82 eV, does not interfere with the first three N^+ peaks. The primary ion beam is not shown, but has the shape of the first peak with a typical full width at half-maximum of 0.8 eV. The first two large peaks are identified as excitations of the N^+ ions from the $2p^2\,{}^3P$ ground state to the $2p^3\,{}^3D$ state requiring 11.42 eV and $2p^3\,{}^3P$ state at 13.52 eV.[14] The energy location is found by a least-squares fit of a parabola to eleven data points (spaced at 0.1 eV) around the maximum. Energy values thus obtained agree with spectroscopic values[15] to ± 0.02 eV.

The small peak at 16.03 eV is evidence of the presence of metastable ions in the incident beam. Only the $2p^2\,{}^1D$ state and the $2p^2\,{}^1S$ state at 1.89 and 4.06 eV above the ground state have transitions which can contribute to this peak. Ignoring intercombination transitions,[16] the transitions contributing to the 16.03-eV peak are excitations from the $2p^2\,{}^1D$ metastable level to the $2p^3\,{}^1D$ and $3s\,{}^1P$ states at 15.98 and 16.60 eV, respectively, and from the $2p^2\,{}^1S$ level to the $3p\,{}^1P$ state requiring 16.34 eV. Intercombination transitions are ignored because peaks such as those which would be produced at 7.36 eV by $2p^2\,{}^1S \rightarrow 2p^3\,{}^3D$ and at 9.53 eV by $2p^2\,{}^1D \rightarrow 2p^3\,{}^3D$ are not observed in the present data.

In order to determine the relative effect of the $2p^2\,{}^1D$ and $2p^2\,{}^1S$ metastables, a detailed analysis of the 16.03-eV peak was carried out for several

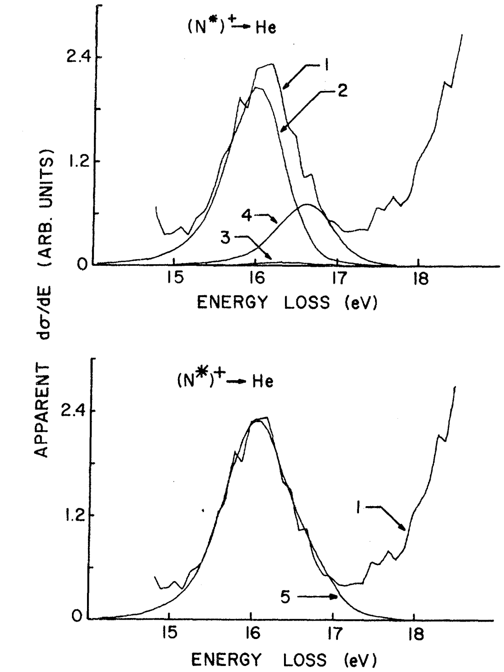

FIG. 2. Detailed analysis of the peak observed at 16.03 eV due to transitions of metastable states only. Curve 1, one data set; curve 2, $2p^2\,{}^1D \rightarrow 2p^3\,{}^1D$; curve 3, $2p^2\,{}^1S \rightarrow 3p\,{}^1P$; curve 4, $2p^2\,{}^1D \rightarrow 3s\,{}^1P$; curve 5, fit to data by adding curves 2, 3, and 4. The data are shown to the same relative scale as that used in Fig. 1.

data sets. Figure 2 shows the results of the analysis for one data set. The procedure used[17,18] (employing a small computer) was to assume the presence of peaks, each having the energy distribution of the primary ion beam, at the spectroscopic locations of the transitions involved. A least-squares fit to the data then yields the appropriate magnitude of each peak which, when summed over all transitions, will reproduce the data. The transition $2p^2\,{}^1S$ to $3p\,{}^1P$ at 16.34 eV (curve 3 of Fig. 2) was given nearly zero amplitude by this process in all data sets analyzed.

In addition to the preceding analysis there is no positive evidence of any of the other transitions which should take place from the $2p^2\,{}^1S$ state. For example, the transition $2p^2\,{}^1S \rightarrow 3s\,{}^1P$ should result in a peak at 14.43 eV, which could be observed even if the magnitude were only a few percent of that for the observed transition $2p^2\,{}^3P \rightarrow 2p^3\,{}^3P$ at 13.52 eV. With the assumption of thermal equilibrium in the source, the population of the $2p^2\,{}^1S$ metastable should be approximately 14% of the $2p^2\,{}^1D$ metastable population and less than 3% of the total beam. Because of these factors, the $2p^2\,{}^1S$ state has been eliminated as a possible beam component in the analysis of the present data.

Normally, the determination of excitation cross sections from energy-loss spectra data is straightforward.[3] However, the presence of the $2p^2\,{}^1D$ metastable state complicates the data analysis. The problem of metastable contamination of ion beams is not new. Techniques previously applied to measure the fraction of metastables[19, 20] are of doubtful value in the present case because of the close energy spacing between the metastable and ground states.

Again, ignoring intercombination transitions, the first two peaks in the energy-loss spectrum are entirely composed of contributions from excitation of the ground state, and the third peak contains only contributions from the metastable state. The energy-loss spectra themselves can then be used to determine the fraction of $2p^2\,{}^1D$ metastables in the beam if this fraction can be made to change between two consecutive observations of the energy-loss spectra. The ion source used is of the discharge type with the discharge maintained between a hot filament and an anode plate. Best operation of the source is at pressures near 50 μm with a discharge of 40 V and about 0.1-A discharge current. Several other modes of operation were tried in order to change the metastable ratio. A higher discharge voltage (near 80 V) did show an increase in metastables.

Writing the ground-state fraction of the N^+ beam as f for the 40-V discharge and as f' for the 80-V discharge, we have

$$\begin{aligned} I_1 &= \sigma_1 f I n l, \quad I_1' = \sigma_1 f' I' n' l, \\ I_2 &= \sigma_2 f I n l, \quad I_2' = \sigma_2 f' I' n' l, \\ I_3 &= \sigma_3 (1-f) I n l, \quad I_3' = \sigma_3 (1-f') I' n' l, \end{aligned} \tag{1}$$

where I is the total incident ion beam current; n the number density of target atoms; l is the collision path length; and I_1, I_2, and I_3 are the ion currents in peaks 1, 2, and 3, respectively, from the cross sections σ_1, σ_2, and σ_3. The primed terms are the same functions for the 80-V discharge. Equations (1) can be readily solved to yield the appropriate ground-state fractions in each of the discharge modes and the cross sections.

The average results of this analysis on several sets of data are shown in Table I. The ratio of the cross sections $2p^2\,{}^1D \to 3s\,{}^1P$ to $2p^2\,{}^1D \to 2p^3\,{}^1D$ in the third peak is given by the curve fitting process (Fig. 2) to be 0.37. This yields estimated cross sections of $(1.2 \pm 1.0) \times 10^{-17}$ cm^2 for the

TABLE I. Experimental results for determination of metastable to ground-state ratio and cross sections.

Parameter	Description	Value	Statistical Error (1 S. D.)	Systematic Error	Error due to f	Total Uncertainty
f	Ground state fraction (40V discharge)	0.90	± 0.05	----	----	± 0.05
1-f	$2p^2\,{}^1D$ fraction (40V discharge)	0.10	± 0.05	----	----	± 0.05
f'	Ground State Fraction (80V discharge)	0.78	± 0.15	----	----	± 0.15
1-f'	$2p^2\,{}^1D$ fraction (80V discharge)	0.22	± 0.15	----	----	± 0.15
σ_1	Cross section for $2p^2\,{}^3P \to 2p^3\,{}^3D$	2.41×10^{-17} cm^2	± 0.23	$\pm 10\%$	$\pm 10\%$	± 0.62
σ_2	Cross section for $2p^2\,{}^3P \to 2p^3\,{}^3P$	1.02×10^{-17} cm^2	± 0.09	$\pm 10\%$	$\pm 10\%$	± 0.27
σ_3	Cross section for $\sum$ $2p^2\,{}^1D \to 2p^3\,{}^1D$, $2p^2\,{}^1D \to 3s\,{}^1P$	1.6×10^{-17} cm^2	± 0.6	$\pm 10\%$	$\pm 50\%$	± 1.1

$2p^2\,{}^1D \to 2p^3\,{}^1D$ transition and $(0.4 \pm 0.3)\times 10^{-17}$ cm^2 for the $2p^2\,{}^1D \to 3s\,{}^1P$ cross section.

The cross sections reported are subject to the assumptions that the cross sections for excitation of intercombination lines from the metastable states are negligible and that the $2p^2\,{}^1S$ metastable fraction of the N^+ beam is negligible. The cross section for the $2p^2\,{}^1D \to 3s\,{}^1P$ excitation could include contributions from the $2p^2\,{}^1D \to 3s\,{}^3P$ transition if these assumptions are not valid. The cross sections for the first two peaks, however, are relatively independent of these assumptions.

The $\pm 10\%$ systematic error given in Table I is an estimate of the uncertainties in pressure, scattering length, and uniformity of source and accelerator over the energy variation of a data trail (50 V out of 50 kV). Possible angular scattering to angles larger than the acceptance angle of the decelerator and energy analyzer has not been included in this error estimate. If angular scattering is significant, the cross sections quoted here will be too low.

No detailed analysis of the remaining structure (Fig. 1) has been attempted. The peaks at 19.2, 21.2, and 23.2 eV are due primarily to excitations of the N^+ ground level, but also contributions from excitation and ionization of He and excitation from the metastable state. The two small peaks between 30 and 35 eV have the correct energy to be simultaneous excitation of the He target and of the first two observed levels of N^+.

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[1]J. T. Park and F. D. Schowengerdt, Phys. Rev. 185, 152 (1969).

[2]J. T. Park, D. R. Schoonover, and G. W. York, Phys. Rev. A 2, 2304 (1970).

[3]D. R. Schoonover and J. T. Park, Phys. Rev. A 3, 228 (1971).

[4]J. T. Park and F. D. Schowengerdt, Rev. Sci. Instrum. 40, 753 (1969).

[5]G. W. York, J. T. Park, J. J. Miskinis, D. H. Crandall, and V. Pol, to be published.

[6]G. R. Burbidge, R. J. Gould, and S. R. Pottasch, Astrophys. J. 138, 945 (1963).

[7]C. B. Tarter, Astrophys. J., Suppl. Ser. 18, 1 (1969).

[8]J. E. Dyson and J. Meaburn, Astron. Astrophys. 12, 219 (1971).

[9]J. W. Chamberlain, *Physics of Aurora and Airglow* (Academic, New York, 1961).

[10]R. R. Brown, J. Atmos. Terr. Phys. 31, 601 (1969).

[11]L. Heroux, Phys. Rev. 153, 156 (1967).

[12]M. F. A. Harrison, K. T. Dolder, and P. C. Thoneman, Proc. Phys. Soc., London 82, 368 (1963).

[13]J. F. M. Aarts and F. J. de Heer, Physica 52, 45 (1971).

[14]Only the term designations [as employed in the work of C. E. Moore, *Atomic Energy Levels as Derived from Analyses of Optical Spectra,* National Bureau of Standards Circular No. 467 (U. S. GPO, Washington, D. C., 1949), Vol. 1] are used here since individual levels of specific J quantum number are not resolved.

[15]Moore, Ref. 14.

[16]J. B. Tatum, Mon. Not. Roy. Astron. Soc. 140, 87 (1968).

[17]D. R. Schoonover and J. T. Park, in *Proceedings of the Seventh International Conference on the Physics of Electronic and Atomic Collisions,* edited by J. B. Hasted (North-Holland, Amsterdam, 1971), p. 839.

[18]G. W. York, Ph.D. dissertation, University of Missouri–Rolla, 1971 (unpublished).

[19]B. R. Turner, J. A. Rutherford, and D. M. J. Compton, J. Chem. Phys. 48, 1602 (1968).

[20]H. B. Gilbody, R. Browning, K. F. Dunn, and A. I. McIntosh, J. Phys. B: Proc. Phys. Soc., London 2, 465 (1969).