Method for measuring the electron excitation cross section of the metastable $1s_5$ level of Ne

Mark H. Phillips, L. W. Anderson, and Chun C. Lin
Department of Physics, University of Wisconsin, Madison, Wisconsin 53706
(Received 29 September 1980)

The electron excitation cross section of the metastable $1s_5$ level of neon is measured by a new technique that involves laser excitation of the metastables to a fluorescing level. A method for obtaining an absolute calibration of the cross section is given. The value of the direct cross section is 16×10^{-19} cm^2 at 20 eV energy of the incident electrons.

In this paper we report the first accurate measurement[1,2] of the absolute direct electron excitation cross section (integrated) of a metastable atom. The metastable levels of an atom are usually relatively low lying and are expected to have large electron excitation cross sections. These cross sections are therefore very important both for their intrinsic interest and for understanding gas discharges such as those used in lasers. The electron excitation cross sections of metastable levels are difficult to measure because these levels do not radiate. Our measurement utilizes a new technique involving laser excitation of the metastable atom to a level that fluoresces. We expect that our technique will be applicable to the measurement of the electron excitation cross sections of the metastable levels of many other atoms and molecules. The energy-level diagram for the processes involved in this experiment is shown in Fig. 1. Neon atoms in the $2p^6\,{}^1S_0$ ground state are excited to the metastable level $2p^53s$, $J=2$ ($1s_5$ in Paschen's notation) by electron impact. This is followed by absorption of 5882-Å laser radiation which takes the metastable atoms from the $1s_5$ to the $2p_2$ ($2p^53p$, $J=1$) level. The subsequent emission from $2p_2$ to $1s_2$ ($2p^53s$, $J=1$) is observed and utilized to determine the electron excitation of the $1s_5$ level.

A schematic diagram of our apparatus is also shown in Fig. 1. A collimated beam of monoenergetic electrons of constant current passes through a collision chamber containing Ne gas at a pressure of 2.5 mTorr and excites the Ne atoms to various energy levels. The fluorescence from the $2p_2$-$1s_2$ transition in Ne is observed at a right angle to the electron beam.[3] The current from a photomultiplier at the output slit of a 0.5-m monochromator is measured with an electrometer which drives the y input of an xy recorder. The x input records the scanned voltage of the electron gun and hence corresponds to the energy of the electron beam. The expanded beam from a dye laser pumped by an Ar-ion laser passes through the collision chamber at right angles to both the electron-beam axis and the fluorescence observation axis. When the dye-laser wavelength is adjusted to the $1s_5$-$2p_2$ transition in Ne, the $2p_2$-$1s_2$ fluorescence is observed to increase substantially over the fluorescence when the dye laser is tuned slightly away from the $1s_5$-$2p_2$ transition. The dye laser is tuned to the $1s_5$-$2p_2$ transition by the use of the optogalvanic effect in a Ne hollow-cathode discharge.[4] In this paper we show that the difference between the fluorescent

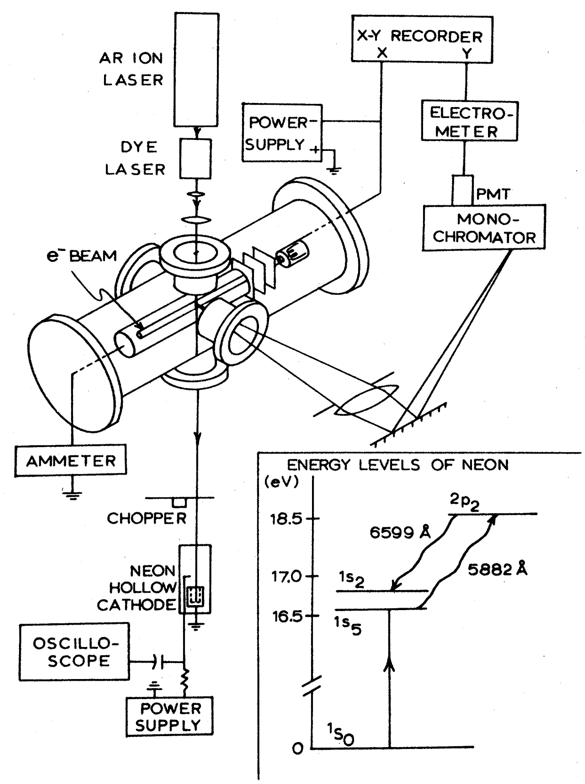

FIG. 1. Schematic diagram of our apparatus and a neon energy-level diagram for the relevant processes.

 ©1981 The American Physical Society

signal with the laser tuned to the $1s_5$-$2p_2$ transition, $S_{\rm on}$, and the fluorescent signal with the laser tuned slightly away from the $1s_5$-$2p_2$ transition, $S_{\rm off}$, is directly proportional to the apparent electron excitation cross section for the $1s_5$ level.

Denoting the $1s_5$ level by a and the $2p_2$ level by b, we write the rate equations for the populations of the two levels:

$$\frac{dn_a}{dt}=n\left(\frac{J}{e}\right)Q_a+n_bA_{ba}+\sum_{\substack{j>a\\ j\neq b}}n_jA_{ja}-n_aA_a-B_{ab}\rho(\nu)n_a+B_{ba}\rho(\nu)n_b \ , \tag{1}$$

$$\frac{dn_b}{dt}=n\left(\frac{J}{e}\right)Q_b+\sum_{j>b}n_jA_{jb}-n_bA_b+B_{ab}\rho(\nu)n_a-B_{ba}\rho(\nu)n_b \ , \tag{2}$$

where n is the atom number density, J is the electron current density, e is electron charge, Q_i is the direct electron excitation cross section for level i, A_{ij} and B_{ij} are the Einstein coefficients, $\rho(\nu)$ is the energy density in the laser beam, and A_i is the reciprocal of the lifetime of level i. Since level a is metastable, A_a is the reciprocal of the beam transit time. The cascade terms in Eqs. (1) and (2) are related to optical excitation cross sections, Q_{ji}, by $n_jA_{ji}=n(J/e)Q_{ji}$. The steady-state solution of Eqs. (1) and (2) gives

$$n_b=\left[\frac{nJ}{e}\left(Q_b^A+\frac{B_{ab}\rho(Q_a^A-Q_{ba})}{B_{ab}\rho+A_a}\right)\right]\Bigg/\left(A_b+\frac{B_{ab}\rho[(g_a/g_b)A_a-A_{ba}]}{B_{ab}\rho+A_a}\right) \tag{3}$$

where Q_a^A and Q_b^A are the apparent excitation cross sections

$$Q_a^A=Q_a+\sum_{j>a}Q_{ja};\quad Q_b^A=Q_b+\sum_{j>b}Q_{jb} \ . \tag{4}$$

The apparent cross section Q_a^A differs from the direct cross section Q_a in that Q_a^A includes both direct production and production by cascading. Note that $Q_{ba}=Q_b^A(A_{ba}/A_b)$. Analysis of n_b requires that we know the magnitude of $B_{ab}\rho$. Our dye laser uses a birefringent filter as the wavelength-selective element. The bandwidth of the dye laser is 4×10^{10} Hz. The longitudinal-mode separation of dye laser is 4×10^{8} Hz. Thus there are about 100 laser modes in the bandwidth of the laser, but usually only one or several modes lase at a given time. As a function of the time the modes that are lasing change so that over a period of time a given mode lases only part of the time. The Doppler width of the $1s_5$-$2p_2$ transitions is 1.4×10^{9} Hz, which is much less than the bandwidth of the laser. Thus even when the laser bandwidth is tuned to cover the $1s_5$-$2p_2$ transition, the laser has for only a fraction of the time a mode lasing that interacts with a segment of the Doppler profile of the Ne atoms. When a laser mode is interacting with a segment of the Doppler profile of the Ne atoms, the intensity in that mode is high enough that $B_{ab}\rho>>A_a$. To see this consider a dye laser power of 100 mW. The photon flux in a 1-mm-diam laser beam is 3.8×10^{19} photons/cm^{2}sec. In the extreme case where all 100 modes are lasing, the flux per mode is 3.8×10^{17} photons/cm^{2}sec corresponding to $B_{ab}\rho\simeq2\times10^{6}$ sec^{-1}. The beam transit time for a Ne atom is such that $A_a\simeq3\times10^{5}$ sec^{-1} so that $B_{ab}\rho>>A_a$. Since the case where all 100 modes lase simultaneously greatly underestimates $B_{ab}\rho$, it is clear that for our experimental situation $B_{ab}\rho>>A_a$ holds whenever a lasing mode interacts with a segment of the Doppler distribution of the Ne atoms. Thus when the laser effect is "on," i.e., one or more of the lasing modes is interacting with a segment of the Doppler profile of the Ne atoms, the b-state population of those atoms is

$$n_{b-{\rm on}}=\frac{nJ(Q_b^A+Q_a^A-Q_{ba})}{e(A_b-A_{ba})} \ , \tag{5}$$

where we have utilized $A_b>>A_a$. When the laser is tuned off of the $1s_5$-$2p_2$ transition or when the laser is tuned to the transition but some of the Ne atoms do not interact with any of the lasing modes, then the population of those atoms is

$$n_{b-{\rm off}}=n(J/e)Q_b^A/A_b \ . \tag{6}$$

The signal of the $2p_2$-$1s_2$ fluorescence when the laser is tuned to the $1s_5$-$2p_2$ transition, $S_{\rm on}$, is proportional to $\beta n_{b-{\rm on}}+(1-\beta)n_{b-{\rm off}}$ where β is a constant determined by a time average over the laser intensity. When the laser is tuned off of the $1s_5$-$2p_2$ transition, the $2p_2$-$1s_2$ fluorescence signal, $S_{\rm off}$, is proportional to $n_{b-{\rm off}}$. The difference $S_{\rm on}-S_{\rm off}$ is proportional to $\beta(n_{b-{\rm on}}-n_{n-{\rm off}})$ which upon using Eqs. (5) and (6) reduces to

$$S_{\rm on}-S_{\rm off}\propto\beta n(J/e)Q_a^A/(A_b-A_{ba}) \ . \tag{7}$$

The fluorescence signals $S_{\rm on}$ at two laser power levels and $S_{\rm off}$ are shown in Fig. 2(a).[5] We obtain Q_a^A from a normalized $S_{\rm on}$-$S_{\rm off}$. For normalization we note that the $1s_5$ level is nearly a pure triplet and

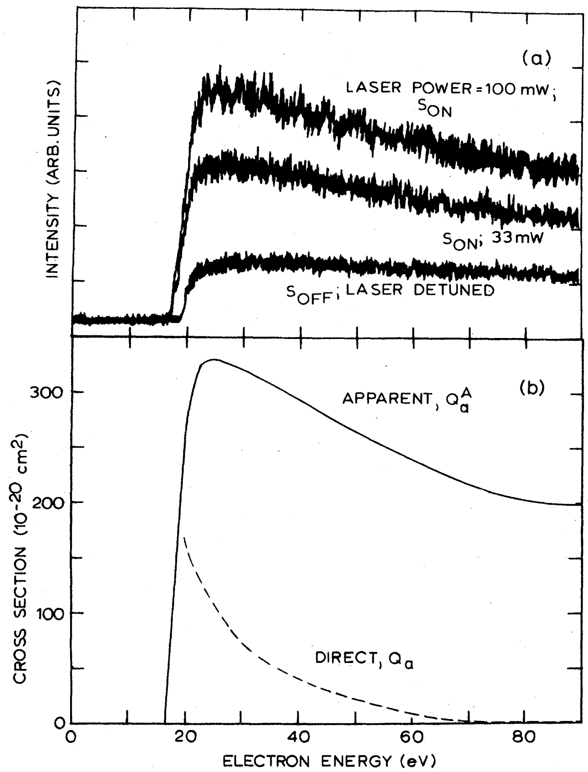

FIG. 2. (a) $S_{\rm off}$ and $S_{\rm on}$ (at 33- and 100-mW laser power) as functions of the electron energy. (b) Apparent excitation cross section, Q_a^A (solid curve) and direct excitation cross section, Q_a (dashed curve) of the metastable $1s_5$ level of Ne as functions of the electron energy.

hence the direct cross section Q_a is expected to peak at an energy slightly above threshold and decrease rapidly with increasing energy. Thus at 90 eV we expect that $Q_a << \sum_{j>a} Q_{ja}$ so that we take $Q_a^A = \sum_{j>a} Q_{ja}$ at 90 eV. Since $\sum_{j>a} Q_{ja}$ has been measured by Sharpton *et al.*,[6] we obtain the calibration of Q_a^A. To obtain Q_a we form the difference $Q_a = Q_a^A - \sum_{j>a} Q_{ja}$. The values of Q_a^A and Q_a as functions of the energy are shown in Fig. 2(b). The cross section Q_a is 16×10^{-19} cm^2 at 20 eV. This is more than six times as large as the peak cross section for the $2s_5$ level as estimated from the data of Ref. 6. We estimate that experimental uncertainty in the energy dependence of Q_a^A is ±10% and the uncertainty in the absolute value of Q_a^A is ±20%. Since $Q_a = Q_a^A - \sum_{j>a} Q_{ja}$, the uncertainty in Q_a is due both to the uncertainty in Q_a^A and the uncertainty in $\sum_{j>a} Q_{ja}$. The uncertainty in Q_a is ±25%.

Several points support our measurements and their interpretation. First, the energy dependence of $S_{\rm off}$ agrees well with that of the apparent cross section Q_b^A measured by Sharpton *et al.*,[6] and the energy dependence of $S_{\rm on} - S_{\rm off}$ for electron energies greater than 65 eV agrees well with that of $\sum_{j>a} Q_{ja}$ measured by Sharpton *et al.*[6] Second, the measured energy dependence of $S_{\rm on} - S_{\rm off}$ is almost independent of the laser power over a range of powers from 30–100 mW. Third, the value of β is expected to vary with the laser intensity as $\sqrt{I}$ since the width of the hole burned in the Doppler distribution by a laser mode is proportional to $\sqrt{1 + I/I_s}$ and since $I >> I_s$ for our experiment where I_s is the saturation intensity.[7] The ratio of β at 100-mW laser power to β at 33-mW power is 1.79 ± 0.08.

In conclusion we report the use of a new experimental technique to make the first accurate measurement of the electron excitation cross section of a metastable level of an atom, the $1s_5$ level of Ne. The large size of the electron excitation cross section indicates the great importance of the electron excitation cross sections of metastable atoms in understanding fundamental processes.

This work was supported in part by the Air Force Office of Scientific Research. We acknowledge helpful discussions with A. R. Filippelli, J. O. Phelps, Dr. J. W. Parker, and Professor J. E. Lawler. We wish to thank Dr. A. W. Johnson for his suggestion of using the optogalvanic effect to set the laser wavelength.

[1]K. Tachibana and A. V. Phelps have measured the excitation coefficient of the metastable $1s_5$ level of Ne and are reporting this measurement at the 1980 Gaseous Electronics Conference. It will be interesting to use our measured cross sections as one input in calculating the excitation coefficient and compare with the results of Tachibana and Phelps.

[2]T. Hadeishi, Ph.D. thesis (University of California, Berkeley, 1962) (unpublished). This thesis reports a measurement of the excitation cross section of the $1s_5$ level by absorption. The measurements were made at 0.4–1.7 Torr where secondary processes may occur. His measured cross sections depend markedly on the pressure and are more than 10 times smaller than the cross sections we report in this paper.

[3]For a discussion of the experimental method for determining electron excitation cross sections of nonmetastable states see R. M. St. John, *Method of Experimental Physics* (Academic, New York, 1969), Vol. 8, p. 27; K. G. Walker and R. M. St. John, Phys. Rev. A 6, 240 (1972).

[4]K. C. Smyth and P. K. Schenck, Chem. Phys. Lett. 55, 466 (1978).

[5]Both $S_{\rm off}$ and $S_{\rm on}$ are observed to depend weakly on the polarization of the fluorescence, and $S_{\rm on}$ is observed to depend weakly on the polarization of the laser. We estimate that polarization effects change the measured cross section by less than 10%.

[6]F. A. Sharpton, R. M. St. John, C. C. Lin, and F. E. Fajen, Phys. Rev. A 2, 1305 (1970).

[7]A. Yariv, *Quantum Electronics*, 2nd ed. (Wiley, New York, 1967).