ELECTRON IMPACT EXCITATION OF COPPER ATOMS

Final Report

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Abstract

The optical excitation function method has been used in a crossed atom and electron beam arrangement to measure the electron impact cross section of the copper $4^2P \rightarrow 4^2S$ resonance lines (324.8, 327.4 nm) from threshold (3.8 eV) to 8 eV. Relative experimental cross section data are normalized at an energy of 1000 eV with respect to first Born theory that includes the $4^2S \rightarrow 4^2P$ resonance transition with an oscillator strength of 0.652 and cascading from the $(3d^{10}nd)^2D$ states with $n = 4, \dots, 10$. The measured Cu $4^2S \rightarrow 4^2P$ cross section is compared with recent theoretical calculations in close-coupling approximation. Very good agreement is found with the ten-state close-coupling theory of Scheibner (private communication).

We have measured the angular-integrated cross section for excitation of the copper $(3d^{10}4p)^2P^o$ state from the $(3d^{10}4s)^2S$ ground state by electrons in the energy range from threshold (3.8 eV) to 8 eV. Low energy electron impact excitation of the $(3d^{10}4p)^2P^o$ state (Fig. 1) is the dominant process for population inversion in the copper vapor laser [1]. Electron collisions in a vaporized copper/neon discharge promote the 4s valence electron of atomic copper to either one of the $J = 1/2, 3/2$ levels of the $(3d^{10}4p)^2P^o$ state. Since this is an electric-dipole allowed transition, this excitation mechanism is rather effective. On the other hand, electron impact excitation of the $J = 3/2, 5/2$ levels of the metastable $(3d^94s^2)^2D$ state is weak since the transition $(3d^{10}4s)^2S \rightarrow (3d^94s^2)^2D$ is strictly forbidden on parity grounds. As the decay of the $(3d^{10}4p)^2P^o$ excited state to the $(3d^{10}4s)^2S$ ground state via the 324.8 and 327.4 nm resonance lines is prohibited by strong radiation trapping at high copper vapor pressure (0.1 Torr at 1500 °C), a three-level system for pulsed laser operation is created.

Copper vapor lasers combine high power (up to 100 W commercially), high efficiency (overall wall-plug efficiency up to 1 %), and high repetition rate (up to several kilohertz) in one single laser system with wavelengths in the green (510.6 nm) and yellow (578.2 nm) portion of visible light. Applications are therefore abundant. The most important single application for copper vapor lasers is in pumping dye lasers. Large-scale laser isotope separation of ^{235}U (AVLIS Project) and ^{239}Pu (SIS Project) at Lawrence Livermore National Laboratory and photoradiation therapy of cancer are outstanding examples. Other applications include detection of trace evidence for law enforcement, underwater transmission, semiconductor processing, and high speed photography [2].

Given the wide field of established and potential uses for copper vapor lasers, it is surprising that very little reliable experimental information exists for the $\text{Cu } 4^2S \rightarrow 4^2P$ electron impact cross section, especially at low electron energies (~ 5 eV) that correspond to the electron temperature of the vaporized copper/neon discharge. Figure 2 summarizes previous experimental work in comparison with two recent calculations. Apparently, the very first experiment was carried out by Williams and Trajmar [3] in 1974 at Jet Propulsion Laboratory who measured differential elastic and inelastic cross sections, integrated the elastic differential data, normalized these integral elastic cross sections to

theory available at the time, and then used measured intensity ratios to obtain integral inelastic cross sections. Their results, $87.8 \pi a_0^2$ at 20 eV and $41.5 \pi a_0^2$ at 60 eV, are *not* shown in Fig. 2, since they have been superseded by another measurement of this group. Trajmar, Williams and Srivastava [4] in 1977 used a similar technique of measurement and data analysis but normalized to different theoretical data. In 1985, Msezane and Henry [5] suggested that the data of Trajmar, Williams and Srivastava should be multiplied by an overall energy-independent normalization factor of 0.36. Shortly after, Msezane and Henry [6] published results of a four-state close-coupling approximation including the $(3d^{10}4s)^2S$ ground state and the $(3d^94s^2)^2D$, $(3d^{10}4p)^2P^o$, and $(3d^{10}4d)^2D$ excited states. They concluded that the measurement of Trajmar, Williams and Srivastava disagreed with their theory both in shape and magnitude. Very recently, Teubner [7] has obtained the integral inelastic $Cu\ 4^2S \rightarrow 4^2P$ electron impact cross section at 60 eV by integrating differential experimental data. His result of $6.0 \pm 0.6 \pi a_0^2$ is in excellent agreement with the value of $6.44 \pi a_0^2$ calculated by Msezane and Henry [6]. Also shown in Fig. 2 is the result of one previous experiment by Aleksakhin *et al.* [8] that used optical detection of the 324.8 and 327.4 nm resonance lines (Fig. 1) to obtain the angular-integrated $Cu\ 4^2S \rightarrow 4^2P$ electron impact cross section. Their data, $14.0 \pi a_0^2$ at 12 eV and $10.7 \pi a_0^2$ at 30 eV, are given with a relative error of $\sim 30\%$. It is obvious from Fig. 2 that the experimental situation with regard to the $Cu\ 4^2S \rightarrow 4^2P$ electron impact cross section is indeed unsatisfactory. This is especially true for the low energy region, say, $E < 8$ eV, where Scheibner, Hazi, and Henry [9] have recently presented a detailed theoretical scattering study based on the four-state close-coupling approximation of Msezane and Henry [6], and where reliable experimental data have never been obtained, inspite of the fact that it is the low energy region that is important for optimized copper vapor laser design.

We have therefore carefully measured the $Cu\ 4^2S \rightarrow 4^2P$ electron impact cross section at low energies. The optical excitation function method described previously [10] was employed: The intensity of the 324.8 and 327.4 nm resonance lines, normalized to unity electron current, was measured as a function of electron energy. The anisotropy of the impact fluorescence was checked and found to be very small. A correction, not carried out here, would affect our data by no more than a few percent.

Thus the observed current-normalized light intensity is directly proportional to the total, that is cascade including cross section $Q_T(4^2S \rightarrow 4^2P)$. Calibration in absolute units of πa_0^2 is accomplished by extending these measurements into the energy range 100 – 1000 eV, confirming that Q_T behaves asymptotically as $\lg_{10}(E)/E$, and then normalizing to first Born theory at 1000 eV. We calculated the direct first Born $Q(4^2S \rightarrow 4^2P)$ using the method described by Krishnan and Stumpf [11] and scaled it to the recommended [12] experimental oscillator strength 0.652. The result is $Q(4^2S \rightarrow 4^2P) = 0.813 \pi a_0^2$ at 1000 eV. We have estimated cascading from the $(3d^{10}nd)^2D$ states with $n = 4, \dots, 10$, again using the method of Ref. 12, and obtained a total cross section $Q_T(4^2S \rightarrow 4^2P) = 0.835 \pi a_0^2$ at 1000 eV to normalize the experimental data. Cascading from the $(3d^{10}nd)^2D$ states thus contributes about 2 – 3 % to the total cross section. No information is currently obtainable for cascading from the $(3d^{10}ns)^2S$ or other higher excited states but it can be expected to be smaller than the already small cascading from the $(3d^{10}nd)^2D$ states. Summing up, we feel that 10 % is a reasonable estimate for the accuracy of absolute calibration.

The apparatus and the experimental procedure have been described in a previous paper on electron impact excitation of 2S and 2D states of rubidium [10]. Briefly, an electron beam of 2.5 mm diameter is crossed with a copper atom beam of 1.2 mm thickness and 5 mm width in a vacuum of 5×10^{-8} Torr. The copper atom beam is produced by a tantalum crucible surrounded by a triple radiation shield machined from 316 stainless steel. A combination of radiation and electron impact heating maintains a crucible temperature of ~ 1100 °C. The electron-impact produced $Cu\ 4^2P \rightarrow 4^2S$ fluorescence at 324.8 and 327.4 nm is collected by a fused-silica f/5 optics, wavelength-selected by an interference filter of 10.3 nm bandwidth, and photon-counted by a cooled photomultiplier (-30 °C). Electron-atom beam overlap and the spatial sensitivity of the photomultiplier cathode are carefully checked.

Typical values for electron current and photon count rate are: At an energy of 4 eV, the electron current passing through the copper beam is $1.8\ \mu A$ and the currents to the entrance and exit apertures, before and after the interaction region, are less than 0.05 % of this current. The photon count rate for

Cu $4^2P \rightarrow 4^2S$ fluorescence is 60 counts/s after subtracting a 1.5 counts/s contribution from oven light.

Our experimental results for the copper $4^2S \rightarrow 4^2P$ electron impact cross section are given in Fig. 3 for the energy range from threshold (3.8 eV) to 8 eV. Individual data points are shown as closed circles with some typical individual error bars, denoting one standard deviation of statistical accuracy. Typically, this is about 5 %. The cross section is observed to rise very rapidly at threshold. We have therefore carefully determined the energy distribution of the electron beam using a retarding potential technique [10]. Results are shown in the insert to Fig. 3: The Faraday current (closed boxes) is measured as a function of a retarding voltage, and an energy distribution (solid curve) with a full width at half maximum of 0.25 eV is obtained by differentiation. Thus the cross section rises from zero to a finite level within an energy range comparable to the energy width of the electron beam. We therefore show in the main part of Fig. 3 the measured Faraday current from the insert, scaled in height and shifted in energy, as solid curve through the threshold cross section data. The excellent agreement between this curve which represents the convolution of a step function with the experimental energy distribution and the experimental cross section data shows that within the 0.25 eV energy resolution of the electron beam our experimental result for the threshold region of the Cu $4^2S \rightarrow 4^2P$ electron impact cross section is indistinguishable from a step-function onset of the cross section. Consequently, we have determined the experimental threshold as follows: The spectroscopic excitation energies of the Cu $4^2P_{1/2}$ and Cu $4^2P_{3/2}$ states [13] are averaged according to an oscillator strength ratio of 2.02 [14] to obtain a spectroscopic $4^2S \rightarrow 4^2P$ threshold of 3.807 eV. The energy where the experimental cross section reaches half of its finite threshold level is then set equal to this spectroscopic threshold value, and an error of 0.1 eV is estimated for the calibration of the energy scale in Fig. 3.

Our experimental results are compared in Fig. 3 with two recent theoretical calculations. The already mentioned four-state close-coupling calculation by Scheibner, Hazi and Henry [9] is shown as long-dashed curve. Very recently, Scheibner [15] has carried out a ten-state close-coupling approximation, adding to the aforementioned four states the six states 4^1P^o , 4^1F^o , 4^1D^o , 2^1F^o , 2^1P^o , and 2^1D^o of the $3d^94s(3D)4p$ configuration. His $4^2S \rightarrow 4^2P$ cross section is given in Fig. 3 by the short-

dashed curve. Comparing experiment and theory, we first note that due to the complex atomic structure of copper the theory of Scheibner, Hazy and Henry misses the spectroscopic threshold by 0.2 eV and the theory of Scheibner obtains a threshold which is still 0.1 eV below the spectroscopic value. Both theories, however, agree in the prediction of an extremely steep onset of the cross section at threshold. As discussed above, this is exactly what we find for the experimental cross section. For energies higher than 4 eV, our data clearly support the more recent calculations of Scheibner in preference to the earlier work by Scheibner, Hazy and Henry. The steep peak at 4.1 eV attributed in the latter work to a $(3d^{10}4p4d)^3F^o$ shape resonance does not exist. Also, the peak at 5.9 eV predicted by Scheibner, Hazy and Henry at the $(3d^{10}4d)^2D$ threshold (calculated by these authors to be at 5.93 eV as compared to the spectroscopic value of 6.19 eV) is not found in our experiment. Instead, the experimentally observed cross section reaches a finite level of $5.5 \pi a_0^2$ at an energy of 4.0 eV and increases smoothly to a local maximum of $8.4 \pi a_0^2$ at an energy of 5.15 eV. Following this local maximum the cross section decreases slightly until 5.6 eV and then rises monotonically to a value of $9.5 \pi a_0^2$ at 8 eV. As illustrated in Fig. 3, these experimental findings are in very good overall agreement with the ten-state close-coupling approximation of Scheibner [15]. When comparing our experiment with the theory of Scheibner three points have to be considered: First, theoretically predicted sharp structures are smoothened by the experimental 0.25 eV energy resolution. Nonetheless we clearly find a local maximum at 5.1 eV as predicted by Scheibner. Second, our experimental data include cascading. An estimated direct cross section is indicated in Fig. 3 at 7.05 and 7.95 eV by open diamonds. Third, our measured cross sections are on the average about 7 % larger than Scheibner's calculated values. This is no significant discrepancy since the theoretical results are estimated to be accurate within 10 % and since the normalization of the experimental cross section scale has been estimated above to be accurate also within 10 %.

In conclusion, we have measured the angular-integrated copper $4^2S \rightarrow 4^2P$ electron impact cross section in the energy range from threshold (3.8 eV) to 8 eV. Statistical uncertainty (one standard deviation) is 5 %, and the uncertainty of absolute calibration is estimated to be 10 %. The uncertainty

of the electron energy is 0.1 eV. Our results are in very good overall agreement with recent, yet unpublished ten-state close-coupling calculations by Scheibner [15]. The large ambiguities resulting from previous experimental work are removed. In order to improve our present understanding of low-energy electron impact excitation of copper even further, many-state close-coupling calculations should be carried out for at least the lower $(3d^{10}ns)^2S$ and $(3d^{10}nd)^2D$ states to enable more accurate cascade corrections at low energies, and first Born calculations, accurate to a few percent, should be carried out for these states to enable a more accurate calibration of experimental data at high energies.

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FIGURE CAPTIONS

FIG. 1: Low-lying energy levels and transitions of copper.

FIG. 2: Previous experimental data for electron impact excitation of the copper 4^2P state in comparison with recent theoretical calculations.

Experiment:

- ◆ — : Trajmar, Williams and Srivastava, Ref. 4;
- ◇ -- : Trajmar, Williams and Srivastava as renormalized by Msezane and Henry, Ref. 5;
- ▲ : Teubner, Ref. 7;
- : Aleksakhin *et al.*, Ref. 8;

Theory:

- : Msezane and Henry, four-state close-coupling calculation for $E > 8$ eV, Ref. 6;
- : Scheibner, Hazi, and Henry, four-state close-coupling calculation for $E < 8$ eV, Ref. 9;

The box in the lower left corner of the figure marks the frame of Fig. 3.

FIG. 3: Low energy electron impact excitation of the copper 4^2P state. The four-state close-coupling calculation (— — —) of Scheibner, Hazi, and Henry (Ref. 9) and the ten-state close-coupling calculation (— — — — —) of Scheibner (Ref. 15) are compared with the present experimental results (●). Typical error bars indicating one standard deviation statistical uncertainty are given. Correction for cascading from $(3d^{10}nd)^2D$ states with $n = 4, \dots, 10$ is indicated (◇). The solid curve through the experimental data at threshold represents the convolution of a step function with the measured electron energy distribution. The insert shows the electron energy distribution with a FWHM = 0.25 eV obtained by a retarding-potential technique (see text).

Figure 1

Figure 2

Figure 3

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