Near-Threshold Measurements of the Spin Dependence of Electron-Impact Ionization

M. H. Kelley, W. T. Rogers,[a] R. J. Celotta, and S. R. Mielczarek
National Bureau of Standards, Washington, D. C. 20234

(Received 20 September 1983)

The spin dependence of the electron-impact ionization of Na has been measured up to 2 eV above threshold with high precision to search for the existence of characteristic oscillations which would support the Coulomb-dipole theory of threshold ionization. The authors have been unable to observe any statistically significant oscillations. The present results are fully consistent with the Wannier theory.

PACS numbers: 34.10.+x, 34.80.Dp

Recently there has been extensive discussion[1] of the relative merits of the Wannier[2-4] and Coulomb-dipole[5] theories of electron-impact ionization of atoms. It has been suggested[6] that a study of the spin dependence of the threshold ionization process might reveal oscillations characteristic of the Coulomb-dipole theory. The Wannier theory, as extended[7] to include spin, predicts a spin dependence which does not vary with energy near threshold. We have therefore carried out a measurement of the energy variation of the spin dependence for near-threshold ionization of Na with high precision and with higher electron-energy resolution than previous experiments.

Briefly stated, the Wannier theory holds that the most probable two-electron escape occurs with the two electrons taking a highly correlated escape route, each having the same energy and maintaining an equal distance from and remaining on opposite sides of the ion core. The threshold law is $\sigma \propto E^{1.127}$, where E is the energy above threshold, and E is uniformly distributed between the exiting electrons. Alternatively, the Coulomb-dipole theory sees the ionization process as dominated by situations where the outgoing electrons have very different energies with the slower electron seeing the charge of the nearby ion core while the faster electron sees the dipole potential of the ion core and the inner electron. The latest treatment of this theory[5,6] includes a nonstatic dipole moment and predicts a modulated, quasilinear threshold law, $\sigma \propto E(\ln E)^{-2}[1+\alpha^{-1} \times \sin(\alpha \ln E + \mu)]$, where α and μ are parameters of the theory. An outgoing electron does not have a uniform probability of having any energy from zero to the excess energy above threshold. This probability oscillates rapidly with energy as energy sharing becomes asymmetric ($\epsilon_1 \simeq 4\epsilon_2$) and approaches zero for the case where one electron possesses all of the energy.

So far, experimental studies of threshold ionization have not proven totally decisive in discriminating between these two theoretical approaches. Temkin has suggested[6] that the asymmetry in the ionization cross section due to the spin dependence of the scattering process should provide a means of distinguishing between the two theories. The asymmetry A is defined as $A \equiv (\sigma_s - \sigma_t)/(\sigma_s + 3\sigma_t)$, where σ_s and σ_t represent the ionization cross sections for the atom's valence electron with the incident electron in a singlet or triplet spin state, respectively. The denominator is the total ionization cross section and A spans the range of $+1$ to $-\frac{1}{3}$ representing the singlet-only or triplet-only scattering limits. In the near-threshold region the Wannier theory predicts a uniform, structureless asymmetry as a function of excess energy while the Coulomb-dipole theory predicts[6] that

$$A(E) \equiv \beta + \tfrac{1}{4}(1+2\beta-3\beta^2)\left[\frac{\sin[\alpha_s \ln E + \mu_s]}{\alpha_s} - \frac{\sin[\alpha_t \ln E + \mu_t]}{\alpha_t}\right], \tag{1}$$

where α_s, μ_s and α_t, μ_t are the Coulomb-dipole parameters for the singlet and triplet initial electron states, respectively, and β is the average value of the asymmetry. Discrimination between the two theories depends on the detection of oscillations in the ionization asymmetry in the near-threshold region and the predictions of the Coulomb-dipole theory depend upon estimates for a number of parameters. For example, if we assume $\alpha_s \approx \alpha_t \approx \alpha$, we can use Eq. (1) to estimate the amplitude of the modulation to be $\approx (1+2\beta - 3\beta^2)/\alpha$. If $\alpha = 10.8$, as suggested in Ref. 6, then the amplitude of the modulation will be ≈ 0.12 or an approximately 33% modulation of the sodium asymmetry.[8,9] This experiment tests an observed asymmetry for statistically significant deviations from a straight line.

The experimental arrangement consists of crossed Na atom and electron beams with a collection facility near their interaction region for detecting individual positive ions as they are produced. The positive-ion current is accumulated as a function of incident electron energy and the polarization directions of the electrons and atoms. The electron beam is produced by the now standard GaAs polarized-electron source.[10] The atom beam, collimated from an effusive oven, is polarized via laser optical pumping.[11] Its polarization is monitored with the use of fluorescence techniques. The apparatus, which will be fully described in a subsequent paper,[12] maintains an ultrahigh-vacuum, magnetically shielded environment.

A typical measurement protocol involves numerous sweeps of the energy range of interest, stepping in both ascending and descending order of electron energy, with the electron polarization direction reversed every 0.5 msec. The direction of atomic polarization is fixed for any given run. Runs of the opposite atomic polarizations are compared as a test for apparatus asymmetries. We have established that systematic apparatus asymmetries are insignificant at the present level of uncertainty.

The electron-energy scale zero is determined by a nonlinear least-squares fit of a model function to the observed total ionization cross section. The model is a threshold power law convolved with a Gaussian electron energy distribution. This determines the energy zero to within ± 0.016 eV for all the reported measurements. The fit also gives the width (full width at half maximum) of the Gaussian used to approximate the electron energy distribution. The mean width for the present data was about 0.15 eV, which agrees with retarding-potential analysis of the beam. Also provided is the energy exponent 1.097± 0.17. These parameters were derived in a fit over the energy range extending up to about 0.8 eV above threshold.

We show in Fig. 1 the measured spin-averaged ionization cross section and spin asymmetry as functions of incident electron energy with data taken at 0.05 eV intervals. An arbitrary scale is used along the y axis since it is the shape of the curves that is of primary interest here. Measurements to resolve a discrepancy which exists between previous determinations[8,9] of the asymmetry are not yet complete. The asymmetry

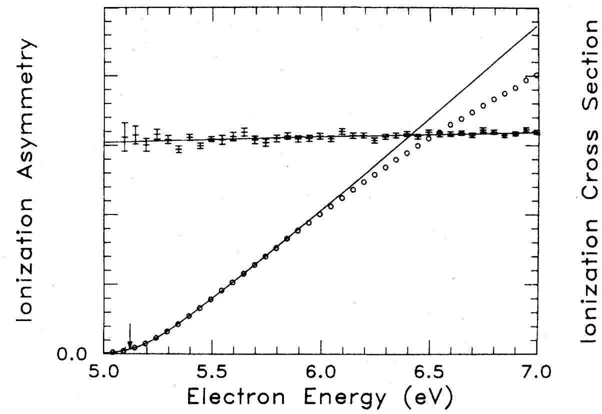

FIG. 1. Measured ionization cross section (circles) is shown with a power-law fit over the range 5.10–5.80 eV. Ionization asymmetry values with ± 1-standard-deviation error bars are shown along with the best linear fit. The threshold energy is indicated by an arrow. Both vertical scales are arbitrary.

data shown represent the summation of thirteen separate data runs. Individual runs consisted of separate measurements of the asymmetry at each energy. A total integration time of 1000 sec per point below 5.5 eV, and 275 sec above 5.5 eV, was achieved. The error bars in the figure represent ± 1 standard deviation in the estimate of the mean of the measurements. These error estimates are based on the counting statistics and agree very well with error estimates derived from the reproducibility of the thirteen independent runs.

The asymmetry data in the figure can be fitted very well with a straight line (also shown) with a reduced χ^2 of 1.014. The slope of the line is not significantly different from zero. Based on a lack-of-fit F test at 5% significance, there is no evidence in the data that a linear function is inadequate to fit the data. Separate fits over the restricted energy range of 5.10–5.80 eV also give excellent fits to a linear function with similar results from the F test. A grouping of data runs with higher energy resolution (0.093 eV), but slightly larger error estimates, is also well fitted by a straight line and shows no significant structure. No statistically significant structure is visible with 1-standard-deviation error estimates of 1.5%–2.5% of the asymmetry. Values of $\alpha = 10.8$ and 40 would imply modulations of 33% and 9% of the asymmetry, respectively. No oscillations of this magnitude are observed.

Our measurements are in full agreement with the Wannier theory's prediction of a uniform asymmetry up to 2 eV above threshold. Further, we agree with the Wannier power-law prediction of the ionization cross section up to about 0.8 eV above threshold with a measured energy exponent of 1.097 ± 0.17. We see no evidence of structures characteristic of the Coulomb-dipole theory. We must conclude that if oscillations characteristic of the Coulomb-dipole theory exist, they must lie outside the range of our current experimental parameters. For example, they may be confined to a region closer to threshold, or vary so rapidly as to not be observable with electron beams of 0.09 eV energy width. Because of the arbitrary nature of the energy scale[13] used in the Coulomb-dipole theory it is difficult to estimate its range of validity or the energy resolution necessary to observe the oscillations. Further experiments with still higher energy resolution will be attempted in the future, but these first measurements support the Wannier theory.

Valuable discussions with A. Temkin are gratefully acknowledged. Research support was provided by the U. S. Department of Energy Office of Basic Energy Sciences.

[a]Present address: Experimental Station, Bldg. 357, E. I. duPont de Nemours & Co., Inc., Wilmington, Del. 19898.

[1]Proceedings of the International Conference on the Physics of Electronic and Atomic Collisions (North-Holland, Amsterdam, to be published), Sessions 5A-1 and 5A-2.

[2]G. H. Wannier, Phys. Rev. 90, 817 (1953).

[3]A. R. P. Rau, Phys. Rev. A 4, 207 (1971).

[4]R. Peterkop, J. Phys. B 4, 513 (1971).

[5]A. Temkin, Phys. Rev. Lett. 49, 365 (1982).

[6]A. Temkin, in Ref. 1.

[7]C. H. Green and A. R. P. Rau, Phys. Rev. Lett. 48, 533 (1983).

[8]D. Hils, W. Jitschin, and H. Kleinpoppen, J. Phys. B 15, 3347 (1982).

[9]G. Baum, M. Moede, W. Raith, and W. Schroder, in Abstracts of the International Symposium on Polarization and Correlation in Electron-Atom Collisions, Munster, 1983 (to be published).

[10]D. T. Pierce, R. J. Celotta, G.-C. Wang, W. N. Unertl, A. Galejs, C. E. Kuyatt, and S. R. Mielczarek, Rev. Sci. Instrum. 51, 478 (1980).

[11]I. V. Hertel and W. Stoll, Adv. At. Mol. Phys. 13, 113 (1977).

[12]M. H. Kelley, W. T. Rogers, R. J. Celotta, and S. R. Mielczarek, to be published.

[13]A. Temkin, IEEE Trans. Nucl. Sci. 30, 1106 (1983).