Experimental evidence for an angular dependence in the ionization probability of silver by 1-MeV protons

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The K-shell ionization probability for silver has been measured at large angles up to 110°. A large increase has been found for the smallest impact parameters which can be explained by a dependence on the scattering angle calculated some years ago by Ciocchetti and Molinari. These theoretical results have been introduced in the binary-encounter approximation (BEA) as an additional correction together with the well-known relativistic, trajectory and binding corrections. In the range of impact parameter considered here, the agreement of this fully corrected BEA with the experimental data is satisfying. New results of the ionization probability for 2-MeV protons on Cu are given; as expected from the theory, the ionization probability does not depend on the scattering angle in this particular case.

I. INTRODUCTION

In the last few years, ionization-probability measurements versus impact parameter have proved to be a very sensitive tool for studying even the finest details of ionization mechanisms. The experimental results can be compared to the semiclassical approximation (SCA)[1] or to the binary-encounter approximation (BEA).[2] In recent reports, Lutz[3] makes an overall comparison of these theories with presently available experimental results. Depending on the combinations of projectile and target, different corrections have to be applied to the basic theories in order to reproduce experimental data. Up to now three effects have been demonstrated to be important in the calculations: relativistic, trajectory, and binding effects.

In the case of 1-MeV protons on gold, Amundsen *et al.*[4] have shown that the one order-of-magnitude difference between theory and experiment can be explained by the relativistic character of the inner electrons. The introduction in the theories of an actual path, combined with the retardation of the projectile in the Coulomb field of the target atom can lead to a decrease of the ionization probability by as much as 50% in the case of 1-MeV protons on silver. The influence of the projectile nuclear charge on the binding energy of the target electron during the collision time was first pointed out by Basbas *et al.*[5] for total-cross-section measurements. The same effect is clearly seen in measurements of the Cu K-shell ionization probability as a function of impact parameter induced by heavy ions.[6] However, the quantitative validity of this correction has not been demonstrated unambiguously. These effects are likely to be more important for low-energy projectiles penetrating deeply inside the K-shell radius of the target atom. Thus, experiments performed at small impact parameters associated with large deflection angles of the scattered particle are expected to provide additional information about the validity of these corrections in the different models.

Furthermore, studying the K-shell ionization probability associated with nuclear reactions, Ciocchetti and Molinari proposed a method to determine the lifetimes of nuclear states.[7] The ionization probability was found to depend on the scattering angle of the emitted particle and on the nuclear lifetime. According to their work, the angular dependence arises from an interference effect between the transition amplitudes associated with the incoming and outgoing particles. In the case of long nuclear interaction time, the interference is destroyed. On the contrary, the anisotropy is found to be a maximum for nuclear interaction times which are equal to zero. Consequently, this effect, first suggested in the nuclear reaction case, should also be present with a maximum amplitude for Coulomb scattering and included in theories describing ionization versus impact parameter.

In this work, we present an extension to large scattering angles for ionization-probability measurements in the case of collision between 1-MeV H^+ and a silver target. We show that the angular dependence suggested by Ciocchetti and Molinari indeed actually exists and leads in this case to an increase of the ionization probability at very small impact parameters which can be accounted for in the very simple BEA theory as an additional correction.

During the development of this work, Andersen *et al.*[8] have performed large-angle ionization-probability measurements on copper at different proton energies. These results show an increase of the ionization probabilities at large angles and low proton velocities, but they contrast with the previous results of Chemin *et al.*[9] in the case of 2-MeV H^+ on copper. In this particular case, we present new results extending up to 90° which are found in good agreement with the data given by Andersen *et al.*[8]

II. EXPERIMENTAL RESULTS AND DISCUSSION

A. Experimental results

The ionization-probability measurements have been obtained by a coincidence technique. Experimental details have been given elsewhere,[9] so we shall only mention the improvements made in the experimental setup.

As the scattering angles become larger, the measurements become more difficult since the counting rate becomes lower, the statistics are poor, and the ratio of true to random coincident events decreases quickly. In order to improve this ratio, a 100-mm²-surface 5-mm-thick Si(Li) diode was used for detecting the K x-rays instead of a 25-mm² surface detector.[9] Also improved was the geometrical disposition of the diode inside the target chamber. In such conditions, the solid angle subtended by the x-ray detector was 0.4 sr, leading to a factor of 10 improvement when compared to the solid angle subtended by the previous detector. In front of the detector, we placed an aluminum foil, 50μm thick, to absorb the Ag L x rays and eliminate the problems related to a high counting rate. Two surface-barrier diodes were used simultaneously to detect the protons scattered at two different angles. Their energy resolution was sufficient to resolve, at backward angles, protons scattered by Ag or Cu from protons scattered by O and C impurities. These corrections for impurities were always very small. The silver and copper targets used were thin (25 μg/cm²) self-supported foils.

The coincidence events recorded on magnetic tape were analyzed with a PDP 15 computer. The time resolution was 12 nsec, and the ratio of true to random events varying from 10 at 30° to 0.9 at 110°.

The experimental results are presented in Figs. 1 and 2 for 1- and 2-MeV protons bombarding silver and copper targets, respectively. In the silver

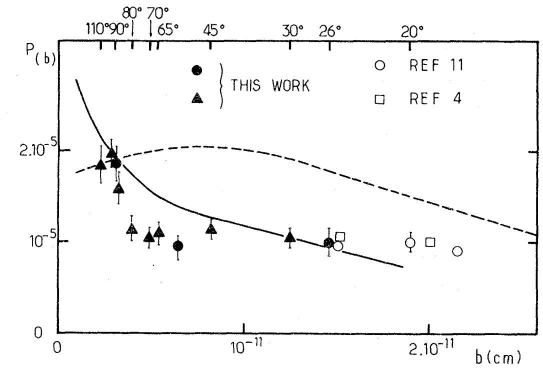

FIG. 1. K-shell ionization probability versus impact parameter in the case of 1-MeV H^+ on Ag. The solid points and triangles represent the results from two independent sets of measurements. The angular aperture of the proton detector was 5°. The theoretical results from the BEA corrected for relativistic, trajectory, and binding effects are given by the dashed line (see text). The full line is obtained from the dashed line when the angular dependence is included as described in the text ($B=-0.4$).

case—Fig. 1—an increase of the ionization probability is clearly seen at large angles. Two independent sets of measurements using different targets during different runs have been performed. Also shown are previous results obtained by Lund[4] and Laegsgaard *et al.*,[11] for smaller scattering angles. In Fig. 2, we show the ionization probabilities measured at 16°, 26°, 31°, 41°, 50°, 65°, and 90° in the case of 2-MeV protons imping-

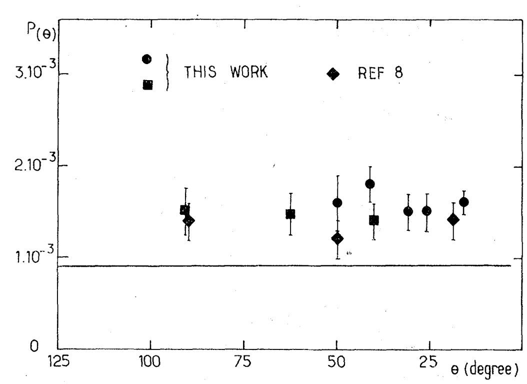

FIG. 2. Dependence of Cu K-shell ionization probability with 2-MeV H^+ on the scattering angle θ. As predicted by the Ciocchetti and Molinari theory, no angular dependence appears in this case ($B=+0.01$). Previous overestimated results obtained at 60° and 36° (Ref. 9) are not plotted, but discussed in the text. The calculated ionization probabilities in the corrected BEA framework are represented by the full line. The comparison between the semiclassical approximation and the binary-encounter approximation has been discussed in the Ref. 9.

ing onto a Cu target with the improved experimental setup described above. Also plotted are the very recent results of Andersen *et al.*[8] up to 90°.

The present results are in close agreement with the data points from Ref. 8, while it appears that the ionization-probability values at 50° and 36° were overestimated by a factor 1.7 and 1.5 respectively, in the previous measurements by Chemin *et al.*[9] The data point at 50° was reproduced several times and the measurements extended to larger scattering angles in order to obtain a significant behavior of the ionization probability at small impact parameters in spite of large uncertainties inherent to this kind of measurement. The discrepancy between Ref. 9 and the present work should be ascribed to the poor efficiency of the small detector previously used leading to very long running time, and poor ratio of true to random coincident events for large scattering angles. Possible systematic deviations are also greatly reduced by detecting protons simultaneously at two different angles.

Finally, it is shown in Fig. 2 that no angular dependence appears in the case of 2-MeV protons impinging onto copper targets.

B. Relativistic, trajectory, and binding effects

In Fig. 3 are plotted the ionization-probability values versus impact parameter obtained from the BEA and SCA formulations in the case of 1-MeV H^+ bombarding a silver target. The corrections applied to BEA have been calculated as described in the following. To determine the trajectory effect, we assume for the projectile a straight-line path tangential to the actual hyperbolic trajectory at the distance of closest approach[1]; the projectile velocity being determined from the conservation-of-energy law at each point. For the relativistic correction, the approximate method indicated by Hansen[10] was used. The change in the binding energy of K electrons due to the presence of the projectile inside the K-shell is accounted for using the relation established by Basbas *et al.*[5] The corresponding modification of the K-shell radius was also introduced.

The comparison of the curves in Fig. 3 indicates the relative importance of the different corrections. From this particular case, several features can be deduced.

Firstly, the magnitude of $P(b)$ obtained from the SCA and BEA theories is in disagreement at the very small impact parameters studied here; the shapes of the curves are also very different at small angles.

Secondly, the magnitude of the various corrections applied to the simple BEA appears to be very large at small impact parameters. The relativistic and trajectory effects change the $P(b)$ value by a factor of 2, but they almost cancel when applied together. Consequently, great care must be taken when comparing experimental and theoretical values, and some agreements found with theories not including all corrections might have been fortuitous, as first pointed out by Burch *et al.*[12] In Fig. 3 are also plotted the curves obtained in the SCA framework taken in the work of Amundsen and Kocbach.[4] Relativistic wave functions and a tangential path are included in the calculations. It must be noticed that the ratio of the corrected values to the uncorrected one at the same angle is quite similar to the SCA and BEA. So, we can be rather confident in the validity of BEA corrections which are quite simple compared to the more elaborated but very cumbersome full SCA calculations.

When all these corrections are applied together we obtain the dashed-line curve in Fig. 1. In a range of impact parameters lying from 0 to 1.5 $\times 10^{-11}$ cm, it is seen that this curve is almost flat. This means that the dependence on impact parameter at large angles is very weak. The same behavior is found from SCA theory in the same range of impact parameter, implying that the ion-

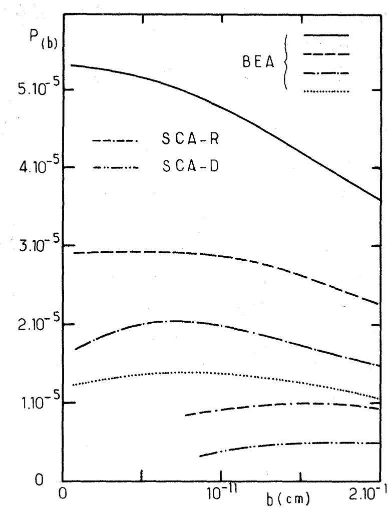

FIG. 3. The set of curves shows the large difference found in the theoretical ionization-probability results depending on the theory used and on the different corrections applied to the simple models. The curves referring to the SCA theory are taken in Ref. 4. The curve labelled SCA-D is corrected for the actual trajectory of the projectile. In the calculation of the curve SCA-R, relativistic wave functions have been included. The dashed line refers to the simple BEA theory. The other BEA curves are calculated with the following corrections; relativistic (full line), trajectory (dotted line), relativistic + trajectory + binding (dot-dashed line).

ization probability does not depend on the geometrical path of the incident projectile as far as we are concerned with atomic regions deep inside the K-shell radius.

C. Interference effect

The ionization amplitude can be seen as the coherent addition of two amplitudes corresponding to the incoming part and the outgoing part of the projectile trajectory. Such a formulation may account for the projectile deflection in a simple way by approximating the actual hyperbolic trajectory by two straight lines separated by the scattering angle θ. An interference effect between the two transition amplitudes appears leading to a dependence of the ionization probability on the scattering angle. This effect was first calculated by Ciocchetti and Molinari, in the case of nuclear elastic scattering and zero-impact-parameter collisions. Such a formulation can be extended to the case of Coulomb scattering and vanishing impact parameter by setting the nuclear interaction time equal to zero in their theoretical expression of the ionization probability. Considering only the partial waves $l=0$ and $l=1$, the Ciocchetti and Molinari expression leads to the following formula for the ionization probability at small impact parameters

$$P(b)=A(1+B\cos\theta), \qquad (1)$$

where b and θ are related by the classical expression.

From a straight-line SCA calculation in the case of 1-MeV H^+ on Ag, the contribution of the $l=2$ partial wave to the ionization probability at 90° is only 0.2% of the contribution of the partial wave $l=0$. Consequently, the $l=2$ and the higher-order partial waves can be neglected.

The angular dependence of the ionization probability on the scattering angle contained in the term $B\cos\theta$ appears when the contribution of the partial wave $l=1$ is considered. The magnitude of this effect is related to the relative importance of the dipole excitation compared to the monopole term and is strongly dependent on the more or less adiabatic character of the collision. According to the calculation of Ciocchetti and Molinari in the elastic scattering case, B is found dependent only on the ratio of the projectile velocity to the K-shell electron velocity, v_p/v_K. For example, they found $B=+0.2$ when $v_p/v_K=0.6$ and $B=-0.4$ for $v_p/v_K=0.2$. This value of $B=-0.4$ corresponds to the case studied here of a collision between 1-MeV H^+ and a silver target, and is used hereafter. When the transition amplitudes associated with incoming and outgoing particles are uncorrelated, which is the case in a nuclear elastic scattering with formation of a long-living compound system, it is shown[7] that the ionization probability becomes twice the one associated with the incoming particle. This is just the value calculated in the BEA formulation. Consequently, we can identify the term A of Eq. (1) with the $P(0)$ value calculated from the corrected BEA. Although this angular-dependence effect is typically a quantum phenomenon which cannot be calculated in the framework of the classical binary-encounter approximation, we feel it is reasonable to introduce it through the theoretical value[7] of B as an additional correction to the simple BEA results. Relativistic and retardation effects are neglected in the calculations of Ciocchetti and Molinari. Nevertheless, it seems that the variation of the anisotropy coefficient B with velocity is not so strong that the neglect of retardation could invalidate our calculation. We assume also that the relativistic effect would not influence the $l=0$ and $l=1$ contributions to the ionization probability in a very different manner.

The final curve obtained by taking into account relativistic, trajectory, binding, and interference effects is represented by a full line in Fig. 1. It is seen that the addition of the present effect leads to a drastic increase of the ionization probability at very small impact parameters. The agreement obtained by the "fully" corrected BEA with experimental data is in fact fairly satisfying in this impact-parameter range.

As previously mentioned, the interference effect is strongly dependent on the velocity ratio v_p/v_K. From the results of Ciocchetti and Molinari, B is equal to 0.01 in a collision involving 2-MeV proton on Cu and consequently, the angular dependence must be very weak in this case. As indicated in Fig. 2, the ionization probability does not change significantly with the scattering angle for θ values varying between 16° and 90°.

Nevertheless, it should be mentioned that the corrected BEA curve is somewhat lower than the experimental data points as shown in Fig. 2.

The dependence of the B term on the velocity ratio is then demonstrated unambiguously by the present results on silver and copper.

III. CONCLUSION

It appears that a complete description of ionization at small impact parameters must include a dependence on the angle of the scattered particle. This effect can only be described by a quantum theory. Nevertheless, it is shown in this paper that this angular dependence at large angle can be introduced as an additional correction to the simple BEA theory together with the now well established relativistic, trajectory, and binding corrections. A good agreement is obtained at *small* impact parameter in the Ag case, considering the absolute values of the ionization probability as well as the dependence on the impact parameter.

Furthermore, it is shown that the comparison between the experimental results and the theories is only valuable if all the "second-order" effects have been carefully investigated.

[1]J. M. Hansteen and O. P. Mosebekk, Nucl. Phys. A 201, 541 (1973).

[2]J. H. McGuire, Phys. Rev. A 9, 286 (1974).

[3]H. O. Lutz, in *Proceedings of the Second International Conference on Inner Shell Ionization Phenomena*, edited by W. Mehlhorn and R. Brenn (Universität Freiburg, Freiburg, 1976), p. 104.

[4]P. A. Amundsen and L. Kocbach, J. Phys. B 8 L122 (1975), and M. Lund, thesis (unpublished).

[5]G. Basbas, W. Brandt, and R. Laubert, Phys. Rev. A 7, 983 (1973).

[6]J. U. Andersen, E. Laegsgaard, M. Lund, and C. D. Moak, Nucl. Instrum. Meth. 132, 507 (1976).

[7]G. Ciocchetti and A. Molinari, Nuovo Cimento 40B, 69 (1965).

[8]J. U. Andersen, E. Laegsgaard, M. Lund, C. D. Moak, and L. Kocbach, J. Phys. B 9, 3247 (1976).

[9]J. F. Chemin, J. Roturier, B. Saboya, Q. T. Dien, and J. P. Thibaud, Phys. Rev. A 11, 549 (1975).

[10]J. S. Hansen, Phys. Rev. A 8, 822 (1973).

[11]E. Laegsgaard, J. U. Andersen and L. C. Feldman, in *Proceedings of the International Conference on Inner Shell Ionization Phenomena and Future Applications, Atlanta*, edited by R. W. Fink *et al*. (Natl. Tech. Information Service, Springfield, Va., 1972), p. 1019.

[12]D. Burch, W. B. Ingalls, H. Wieman, and R. Vandenbosch, Phys. Rev. A 10, 1245 (1974).