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Paul Burrow

J.A. Michejda

J. Comer

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Low-energy electron scattering from Mg, Zn, Cd and Hg: shape resonances and electron affinities

P. D. Burrow,^{1,3} J. A. Michejda,¹ and J. Comer²

¹ Department of Engineering and Applied Science, Mason Laboratory, Yale University, New Haven, Connecticut 06520, USA

² Department of Physics, The University, Manchester M13 9PL, England

³ Department of Physics and Astronomy, University of Nebraska, Lincoln, Nebraska 68588, USA

Abstract

Measurements of electron scattering at low energy from Mg, Zn, Cd, and Hg have been carried out using the electron transmission method. A large shape resonance is observed for each element which we identify with the $(ns^2np)^2P$ ground state of the negative ion. The electron affinities in eV are found to be Mg(-0.15), Zn(-0.49), Cd(-0.33), and Hg(-0.63) with an error of ± 0.03 eV. The results are compared with the available theoretical predictions.

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1. Introduction

The introduction of an electron into the lowest normally unoccupied atomic orbital of most elements results in a stable negative ion. For atoms in certain columns of the periodic table, the rare gases most conspicuously, no bound states of the negative ion are known to exist. It is less certain but now generally agreed that the negative ions of the group II elements are also unstable (Hotop and Lineberger 1975). In this paper we present an experimental study of the electron scattering from certain elements of columns IIa and IIb with a view toward describing the nature of an electron which occupies the lowest normally unfilled orbital.

If the system comprising the atom and electron can be regarded as a negative ion, its energy is greater than that of the neutral atom, and the ion is embedded in the continuum of states of the free electron and atom. The system is thus unstable against autodetachment of the electron and, at best, must be classified as a temporary negative ion. Because of its location in the continuum, such an ion may be formed by electron impact on the neutral atom if the electron energy is judiciously chosen. Under conditions to be described below, the short-lived occupation of an empty orbital may be manifested as a rapid variation with energy of the electron scattering cross section, or "resonance." The reader is referred to the review by Schulz (1973) of such phenomena in electron scattering from diatomic molecules.

Do we expect to observe such resonances in all atoms which do not have stable negative ions? If we require "rapid" variations in the cross section: probably not. Because of their closed-shell configuration, the rare gases present the worst case. An electron in the lowest normally unfilled orbital lies at a large radial distance from the atom and sees little of the polarization potential required to localize the electron wavepacket. Consequently, the lifetime of the negative ion is likely to be so short and the associated variation in the scattering cross section so broad, that the resonance is not easily discernible against the smooth variation with energy of the non-resonant background cross section.

The group II elements, on the other hand, are ideal candidates for sharp resonant structure because of three important characteristics. Firstly, they have an ns^2 outer electron configuration, that is, a filled

subshell. This property most likely prevents a stable negative ion in the same manner as the closed shell in the rare gases. Secondly, the lowest unfilled orbital, for example np in the IIb elements, occurs in the *same* principal quantum number as the highest filled orbitals. The additional electron therefore lies at relatively low energy and experiences a sizable contribution from the polarization potential. Thirdly, and of great importance, an additional electron in an orbital with $l > 0$ sees a centrifugal barrier through which it must tunnel to auto-detach. The combination of these potentials is necessary to localize the electron wavepacket sufficiently to create a resonance observable in an electron scattering experiment. Such a feature is commonly labelled a “shape” resonance in the literature.

In the following sections, we describe an examination of the total electron scattering cross sections of Mg, Zn, Cd and Hg using an electron transmission method. In each case a pronounced resonance peak is observed below 1 eV which we associate with the formation of the ground state of the temporary negative ion. Preliminary reports of portions of the data have been published elsewhere (Burrow and Michejda 1974, Burrow and Comer 1975a, b).

2. Apparatus

The study of electron scattering from metal vapors with good energy resolution and at impact energies below 1 eV presents a number of experimental problems. Because we are concerned with accurately locating sharp structure rather than measuring absolute magnitudes of cross sections or the dependence on electron scattering angle, we have employed the electron transmission method in the format using an axial magnetic field. In addition to being conceptually straightforward, the apparatus is usable even in relatively hostile environments and no unusual difficulties were encountered in adapting it for use with metal vapors.

We first review briefly the idea behind the transmission method and then describe the apparatus in more detail. **Figure 1** illustrates the main components schematically. An electron monochromator injects a beam of current I_0 into a scattering chamber containing gas at density N and total cross section Q . After traversing the pathlength L ,

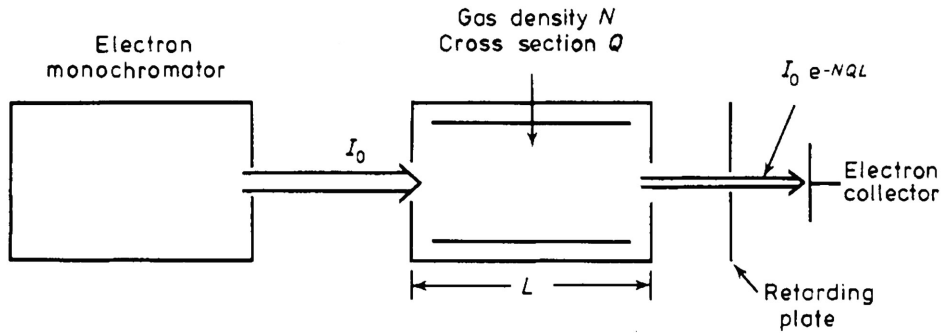


Figure 1. Schematic drawing of electron transmission apparatus.

the scattered portion of the incident beam is rejected by means of an aperture or by discriminating against the reduced axial velocity component at a retarding plate. The current transmitted to the collector therefore is the *unscattered* current given by $I_0 \exp(-NQL)$. Thus a signal is derived which is related to the total scattering cross section as a function of electron energy.

In the present apparatus a magnetically collimated electron beam with currents in the 5×10^{-9} A range and energy resolution of 30 to 60 meV full width at half maximum was produced by a trochoidal monochromator similar to that described by Stamatovic and Schulz (1970). By careful alignment of the apertures with the magnetic field and proper tuning of the monochromator, the transmission properties of the apparatus could be made quite uniform over the range of energy examined here.

Because of the axial magnetic field, typically 150 G, the scattered electrons are constrained to spiral along the field lines together with the unscattered electrons. Although a portion of the scattered current is rejected at the exit aperture of the collision chamber, the discrimination is increased by retarding at an electrode following the collision region. Typically, the unscattered beam energy is reduced to less than 200 meV at the retarding plate. For our purposes precise knowledge of the angular resolution for rejection of the scattered current is not necessary. Appropriate checks were made to ensure that the energies of the features studied here were not dependent on the retarding voltage.

All electrodes in close proximity to the electron beam were constructed of molybdenum. The apertures were positioned axially and

spaced by sapphire balls placed in carefully machined alignment holes.

To study metal vapors, provisions for operation at high temperatures are necessary. Two different configurations of the scattering chamber have been used successfully. The first, suitable for the metals studied in this report, was a standard static gas cell wrapped with ceramic-insulated tungsten wire which was resistively heated. Separate heaters were employed on the gas cell, the reservoir containing the metal under study, and around the metal tube connecting the two. It was also found advantageous to apply heat to the electron gun to reduce deposition of metal on the sapphire spacer balls.

In anticipation of studies at substantially higher temperatures, an alternate collision geometry was tested in Mg, Cd and Zn. This configuration employed a well shielded tantalum oven heated by electron bombardment. The beam of metal atoms emerging from a small hole in the oven was cross-fired with the electron beam at a distance of about 6 mm. Although the effective pathlength for electron scattering is much shorter than in the static cell, no difficulty in achieving a usable attenuation was encountered. The signal-to-noise ratio and long-term stability were superior in the static cell geometry, however, and all the data presented here were taken in this mode.

3. Energy-scale calibration

The results of the present work are displayed below as plots of the transmitted current as a function of the incident electron energy. For the energy-scale calibration, however, the derivative of the transmitted current was recorded by modulating the electron impact energy and synchronously detecting the corresponding AC component of the transmitted current (Sanche and Schulz 1972). This technique enhanced the sharp structure in the transmitted current and made the energy calibration more accurate.

Two separate methods were used to calibrate the energy scale. Although no provision for introducing a calibration gas directly into the collision chamber was made, it was found that by flooding the vacuum envelope with the reference gas to a pressure of approximately 5×10^{-3} , resonances useful for calibration could be observed. In particular, the N_2 resonances in the 2 eV region could be seen easily. These

in turn were referenced to the well known $\text{He}^- (1s2s^2)^2\text{S}$ resonance in a separate apparatus. As our primary standard we took the minimum in the derivative of the transmitted current through helium to be at 19.37 eV (Sanche and Schulz 1972). This positions the $\text{N}_2^- (^2\Pi_g)$ resonance such that the first maximum in the derivative of the transmitted current lies at 1.98 eV.

This calibration procedure was not entirely satisfactory as the N_2 resonances are rather broad and the presence of a relatively high density of N_2 in the region of the monochromator tended to degrade the electron gun characteristics. Additional support for the calibration was obtained with a second procedure which utilized the structure appearing in the derivative spectra close to the lowest excited ^1P state in each atom. This feature, which is narrow and well isolated, is almost certainly a cusp in the scattering cross section due to the opening of the ^1P channel. Such cusps have been previously observed in helium (Sanche and Schulz 1972) and in the alkali metals (Andrick *et al.* 1972) and have been discussed in more detail by Cvejanović *et al.* (1974). That these structures are indeed cusps is supported by two pieces of evidence. Firstly, their shapes are similar to those predicted for cusps and different from typical resonance profiles. Secondly, our calibration using N_2 and He places the centers of the cusps at the energies of the associated ^1P states within 20 meV or less for each gas. Reinforced by this agreement, we estimate the overall error in the energy scale to be less than 0.030 eV. The cusps provide a potentially more accurate means of energy calibration; however a detailed study of the cusp profile and its broadening by the electron energy distribution is required.

We note here that the energy calibration of the resonances discussed later in this paper was carried out at low electron beam attenuation to avoid distortion of the resonance profiles. If $NQL \ll 1$ then the derivative signal is simply proportional to $-dQ(E)/dE$.

4. Results

Figures 2-5 illustrate typical direct transmission measurements in Mg, Zn, Cd and Hg, respectively. In each case the transmitted electron current in arbitrary units is plotted as a function of electron impact

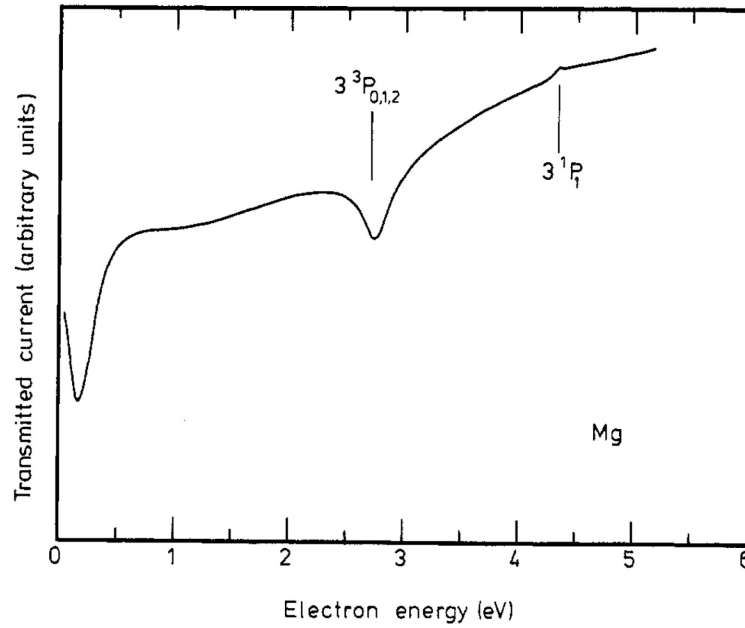


Figure 2. The transmitted current as a function of electron energy in magnesium.

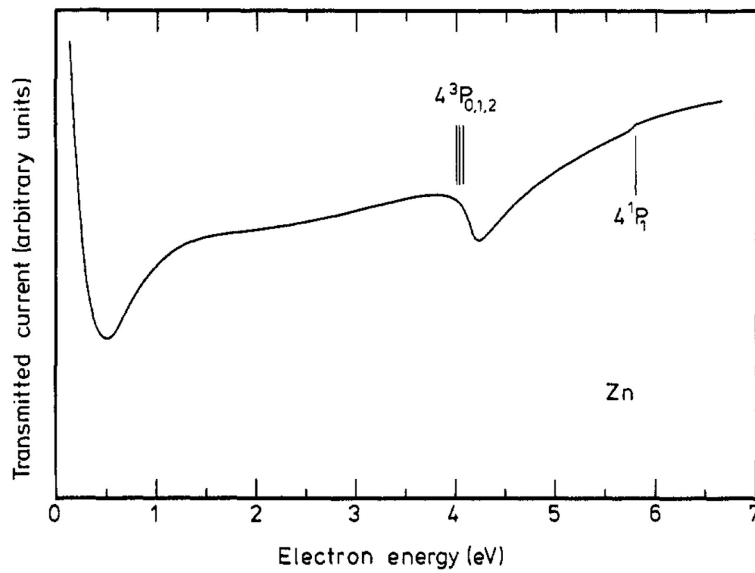


Figure 3. The transmitted current as a function of electron energy in zinc.

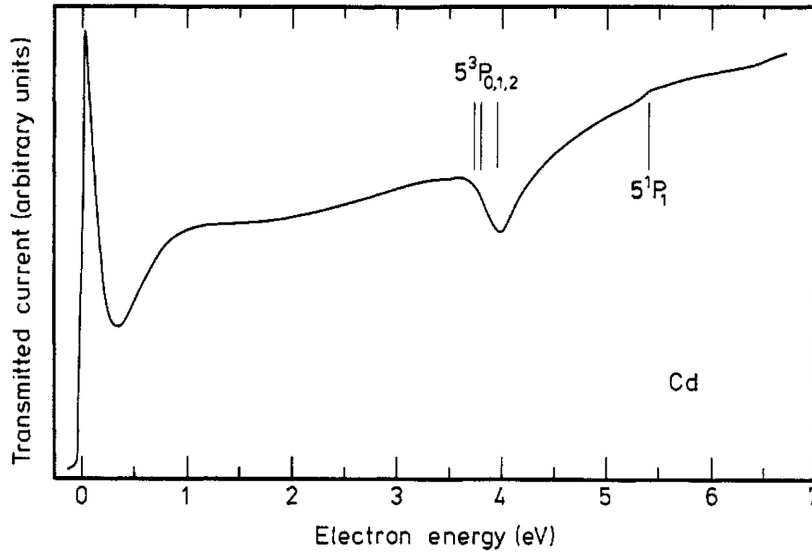


Figure 4. The transmitted current as a function of electron energy in cadmium.

energy. The vertical lines indicate the energies of the lowest multiplet of excited states of the neutral atom. The energy scale in these figures was positioned by adjusting the curves such that the 1P state is appropriately located with respect to the cusp, as determined by our calibration using the derivative method. In the case of Hg, for which the cusp is not evident in the direct curve, the resonances near the 3P states were used as intermediate references.

It is important to note here that the electron beam attenuation was deliberately chosen to be high, $NQL > 1$, in figures 2–5 to increase the visibility of the sharp features for display. Under these conditions, the curves are not suitable for accurate calibration of the low-energy features. In Hg, for which the attenuation was particularly high, the lowest dip in the transmitted current is shifted by about 0.08 eV from its position determined in our low-attenuation derivative measurement.

In Figure 4 the abrupt rise in transmitted current near zero energy is illustrated. The maximum in transmitted current just above zero is critically dependent on the resolution and tuning of the gun. The convolution of the cross section with the partially retarded electron beam energy distribution at absolute energies in the range 0–50 meV causes the transmitted current to be largely instrumental in this region. The onset of beam current in the other figures is deleted.

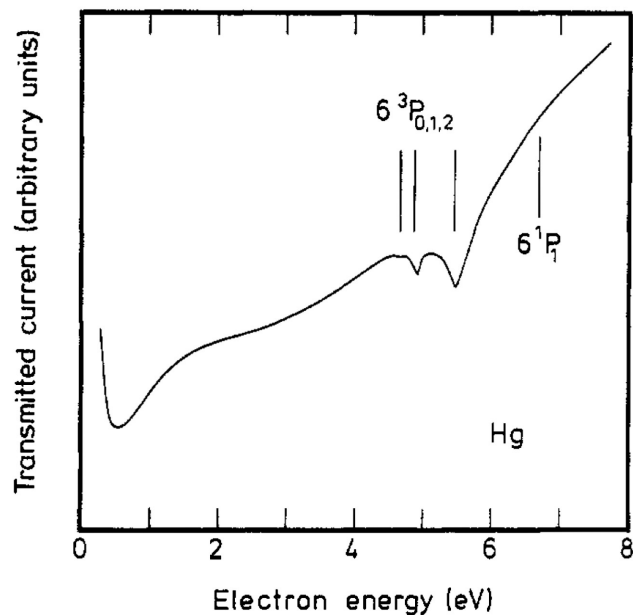


Figure 5. The transmitted current as a function of electron energy in mercury.

Considerable structure in the scattering cross section is apparent over the range of energies displayed here. In the present paper we will be concerned only with that portion of the scattering below the electronically excited states. A report on the remaining features as viewed with the derivative method will be presented at a later date (Burrow and Michejda, in preparation).

Figures 2-5 indicate clearly that electron scattering from each of these group II elements is dominated at energies below 1 eV by a large asymmetric peak in the cross section. Although there is a strong family resemblance in the spectra, as one expects for members of the same column of the periodic table, there is considerable variation in the widths and locations of the low-energy peaks.

There is surprisingly little experimental data in the literature with which to compare these results. A review of electron-atom collision cross sections may be found in Bederson and Kieffer (1971). The early work of Brode (1929, 1930) using the modified Ramsauer technique indicated a monotonically rising cross section with decreasing energy in Zn, Cd and Hg. The experiments were not carried out below about 1 eV however and a maximum in the cross section was not observed. A swarm experiment in Hg was carried out by McCutchen (1958) who

analyzed his results with simple kinetic theory. Unambiguous evidence for a pronounced peak in the cross section was found, although the energy (0.25 eV) is considerably below that located here. More recently Rockwood (1973) has analyzed the existing swarm and transport data in Hg and finds a similar peak in the momentum transfer cross section, the energy of which is in excellent agreement with the present results.

5. Discussion

We attribute the low-lying peak in the cross sections of the four elements examined here to a resonance caused by the temporary occupation by the impacting electron of the lowest unfilled atomic orbital with non-zero angular momentum. The partial wave in which the resonance occurs cannot be determined directly from the present experiment since the signal is related to the total scattering cross section. A measurement of the angular scattering dependence would be required for this purpose. The assignment, however, is straightforward and is supported by the more detailed evidence described below. The adjacent elements of the periodic table which are isoelectronic to the temporary negative ions, for example Al in the case of Mg^- , are built up by the addition of an np electron to the ns^2 subshell. Thus we expect that the ground states of the temporary negative ions will be designated as $(ns^2np)^2P$ where $n = 3, 4, 5,$ and 6 for Mg, Zn, Cd, and Hg, respectively.

The energy with which the additional electron is bound to the neutral atom defines the electron affinity of the neutral. For unstable negative ions the magnitude of the affinity is given by the electron energy at which the resonance in the scattering cross section occurs; the sign is taken by convention to be negative. The determination of the resonance energy from the experimental data requires further comment. In general a resonance profile is described by an expression containing both constructive and destructive components whose admixture is determined by the background phaseshift of the resonant partial wave. Normally a fit of the experimental profile to the theoretical expression is required to extract the relevant parameters. The shape resonances encountered in the present work occur

Table 1. Energy of $(ns^2np)^2P$ negative ion above $(ns^2)^1S$ neutral (eV).

	This work	Zollweg (1969) (Horizontal analysis)	Hunt and Moiseiwitsch (1970) (Model potential)	Robb (unpublished) (Close coupling)
Mg	0.15 ± 0.03	0.52	0.37	0.166
Zn	0.49 ± 0.03	0.67		
Cd	0.33 ± 0.03	0.78 (0.818)*		
Hg	0.63 ± 0.03	0.67 (0.829)*		

* Values in parentheses correspond to the configuration center of the $^2P_{1/2,3/2}$ levels. The remaining values are for the $^2P_{1/2}$ level.

at sufficiently low energies that the background p-wave phaseshift is not expected to be appreciable. This implies that the resonance energy corresponds closely to the energy at which the maximum in the cross section appears.

The experimental resonance energies are listed in **Table 1**. The electron affinity for addition of a p electron to the neutral atom is thus given by the negative of the entry in the table. These values were calibrated using the derivative method described earlier.

5.1. Comparison with theory

A number of theoretical and semi-empirical methods have been used to determine the energy with which the additional electron is bound in a negative ion (Hotop and Lineberger 1975). For unstable negative ions which lie in the continuum of states of the neutral atom and a free electron, the methods of scattering theory are also applicable. In this section we discuss the predictions of these methods which pertain to the present work.

The only study which has been applied uniformly to the elements examined here is that of Zollweg (1969) who employed horizontal analysis, a semi-empirical extrapolation method. The electron affinities derived by this analysis were adjusted to fit accurate experimental values for one reference ion in each horizontal series. The calibration affinities used by Zollweg were those of Cl, Br, and I. To extend the results to the 6p subshell the halogen electron affinities were extrapolated vertically to yield an estimated value for astatine. Although

it is difficult to estimate the accuracy of the results obtained by horizontal analysis, Zollweg suggests that the average error is 0.1-0.2 eV except for the 6p subshell where it may be about 0.3 eV. This estimate appears to be borne out by Hotop and Lineberger (1975) who find that the predicted results for the main group elements of the third, fourth and fifth rows agree to within 0.2 eV or better with the available accurate experimental determinations.

The predicted energies for the ${}^2P_{1/2}$ state of the negative ions as derived by Zollweg (1969) are listed in Table 1. It must be noted that for the heavier elements there is substantial spin-orbit splitting between the ${}^2P_{1/2}$ and ${}^2P_{3/2}$ levels. In Hg this amounts to 0.238 eV and in Cd, 0.057 eV, as determined by Zollweg from quadratic extrapolation of isoelectronic series. In the present experimental study there is no indication of doublet formation in the low-lying resonance. Indeed, since the spread in energy due to the finite lifetime of the negative ion is comparable to the predicted spin-orbit splitting, it is not clear that the doublet structure has time to develop. It may be more appropriate therefore to compare the experimental results with the predicted energies of the ${}^2P_{1/2,3/2}$ configuration center. These values are also listed in Table 1. The average overall difference in the predicted configuration centers of the four elements and our experimental results is about 0.3 eV, which would appear to be acceptable agreement. The extrapolated values are uniformly higher than those measured here.

The remaining comparisons with theory are discussed individually.

5.2. Magnesium

Magnesium, being the lightest element studied here, is the most tractable system for theoretical study. Hunt and Moiseiwitsch (1970) have carried out a calculation of e-Mg scattering in the p partial wave using a model potential. One adjustable radius parameter was incorporated which was determined by requiring the potential to yield the experimentally observed energies of a given state of the isoelectronic positive-ion sequence. A quadratic extrapolation of the parameter to the isoelectronic negative ion was then made. The resulting potential was used to model the field experienced by the electron in scattering from the neutral atom. A pronounced p-wave resonance was located associated with formation of the 2P state of e + Mg at 0.37 eV. The

corresponding resonance width, Γ , was 0.1 eV. A comparison with experiment of the affinities computed by Hunt and Moiseiwitsch for stable negative ions yields agreement within 0.1 eV for most cases. The result for the Mg shape resonance lies above our experimental value by 0.22 eV but is in substantially better agreement than the value determined by horizontal analysis.

Two close-coupling calculations of e-Mg scattering have been published (Van Blerkom 1970, Fabrikant 1974) with conflicting results at low energy. Both calculations appear to be superseded by recent work of W. D. Robb (unpublished) who finds a p-wave shape resonance at 0.166 eV, well within our experimental error limits of ± 0.030 eV.

5.3. Mercury

Following the methods of Bransden and McDowell (1969), Hutt (1975) has recently carried out a phaseshift analysis of e-Hg scattering at energies below the electronically excited states. In this procedure all of the experimental data are fitted simultaneously with a consistent set of phaseshifts for the important low-order partial waves. The phaseshifts are parametrized as continuous functions of energy. In the latest study the momentum transfer cross section derived by Rockwood (1973) was included along with other recently revised data and a much more satisfactory fit was made than in the previous attempt (Hutt and Bransden 1974). Of particular importance to this paper was Hutt's result that the best agreement was obtained when the $p_{1/2,3/2}$ phaseshifts were parametrized in explicitly resonant forms in the region of the peak in the cross section.

In spite of the manifest theoretical difficulties involved in an understanding of e-Hg scattering at low energy, Walker (1975) has carried out a numerical solution of the Dirac equation including exchange and a polarization potential. He finds a pronounced resonance in the $p_{1/2}$ partial wave at approximately 0.2 eV. A much broader resonance in the $p_{3/2}$ partial wave was also located whose characteristics were somewhat dependent on the degree of exchange included. It is not yet clear whether the $p_{3/2}$ resonance will appear as a discernible second peak or only a broad variation in the cross section which is indistinguishable from the non-resonant contributions. On the basis of his calculations, Walker favors the latter alternative.

Although neither of these studies provides data which are detailed enough to compare directly with our measurements, they yield strong evidence to support our original suggestion of the resonant p-wave.

As mentioned earlier, the momentum transfer cross section derived by Rockwood (1973) from an analysis of swarm and transport data contains a peak at 0.625 eV with which our results are in excellent agreement.

5.4. Shape resonance characteristics

Certain systematic characteristics of the p-wave shape resonances are worth further comment. In **Table 2** we have listed the elements in order of increasing resonance energy. In the second column we have entered a crude measure of the inverse lifetime of the resonance, namely, the width of the resonance peak. The particular values given are the spacing between the extrema in the derivative of the transmitted current. These measurements were made at very low electron beam attenuation. The quoted widths therefore are the spacing between the energies at which the maximum positive and negative slopes of $Q(E)$ occur.

Table 2 clearly indicates that the lower the resonance energy, the sharper the peak, or alternatively, the longer the lifetime of the temporary negative ion. This is a well understood characteristic of shape resonances, as illustrated for example by Bardsley *et al.* (1967). Recall that in a simple picture the potential seen by the electron can be broken into two primary components, the short-range shielded Coulomb hole and the much more extended centrifugal barrier. The lifetime of the temporary negative ion is chiefly determined by the height and radial extent of the barrier through which it must tunnel. The lower the resonance energy, assuming the barrier characteristics are unchanged, the greater the portion of the barrier seen by the electron

Table 2. Energy and width of $(ns^2np)^2P$ resonances.

	Energy (eV)	Width (eV)
Mg	0.15	~0.14
Cd	0.33	~0.33
Zn	0.49	~0.45
Hg	0.63	~0.4

and hence, the longer the ion lifetime. Each of the atoms in table 2 has a p-wave barrier, thus this part of the potential seen by the electron is not altered. In passing through the various elements, we vary, crudely speaking, the shielded Coulomb binding responsible primarily for the resonance energy. Thus we expect a monotonic variation of width with energy as illustrated in Table 2. We hasten to add, however, that a detailed understanding of the lifetimes will require taking into careful account the relative polarizabilities and sizes of these atoms as discussed briefly by Bardsley *et al.* (1967).

In contrast to the closed-channel Feshbach resonances observed in atoms (Schulz 1973), the energy spread of the shape resonances is considerably greater than that of the electron beam. The resonance may therefore be "excited" at different energies within its profile. On the low-energy side the effective barrier seen by the electron is more substantial than on the high-energy side. Thus it appears that the resonance lifetime is energy dependent (Henry *et al.* 1969). This is borne out by the asymmetric profiles observed here and in shape resonance calculations (Hunt and Moiseiwitsch 1970, Henry *et al.* 1969). The resonance width at the peak of the cross section, that is at the resonance energy, therefore represents an intermediate value over the whole resonance given only approximately by the width at half maximum of the peak. A careful fit to the data with a model profile would be necessary to extract the actual width and its energy dependence.

5.5. Other negative-ion states

It is appropriate to discuss briefly other states formed by addition of an electron to the ground electronic state of the group II atoms. In Mg, Zn, Cd, and Hg, quadratic extrapolation of the isoelectronic series indicates that the $(ns^2(n+1)s)^2S$ states are more stable than the $(ns^2np)^2P$ state discussed here but not bound with respect to the neutral atom (Zollweg 1969 and references therein, Weiss 1967). If such a state lies in the continuum, it will not be observable with the present technique. Having no angular momentum barrier to trap it, the s electron will not be localized on the atom long enough to create a resonance. The effect on the cross section will be indistinguishable from the background non-resonant scattering. An exception to this might

occur if the state appeared just above zero energy. Such virtual states are discussed in the review by Taylor (1970).

We must also consider the possibility that contributions to the total scattering cross sections are made by shape resonances with $l > 1$. The most likely candidate is a temporary negative ion in the $(ns^2nd)^2D$ state. A crude estimate of the location of this state, in Mg^- for example, can be obtained from an isoelectronic extrapolation of the corresponding levels in Al I, Si II, P III, and S IV. Such an extrapolation was carried out for each of the four elements studied here and, although the extent to which such a procedure can be trusted is not clear, it predicts that the $(ns^2nd)^2D$ state lies above the $(ns^2np)^2P$ by 1 eV or less. There is faint evidence for a very broad feature in each cross section between the p-wave shape resonance and the first excited states of the neutral. It is tempting but entirely speculative to suggest that this is associated with a d-wave shape resonance. A measurement of the angular scattering dependence in this region or a detailed theoretical examination of the phaseshifts might shed some light on this question. Because of the great breadth of this feature, it is unlikely that the d-wave phaseshift will advance by π radians; classification as a resonance therefore may not be useful.

Although the ions Zn^- , Cd^- , and Hg^- have been observed by Kaiser *et al.* (1971) in a mass spectrometer, it is generally believed that these are metastable species corresponding to doubly-excited states of the negative ion (Hotop and Lineberger 1975).

6. Concluding comments

The four elements studied here, along with the rest of the IIa elements, are unique in that they apparently do not possess stable negative ions. The existence of the ground state of the negative ion is therefore manifested as a large peak in the total electron-atom scattering cross section at low energy. Such resonances, however, are not restricted to these elements. Similar structure will appear in electron scattering from elements in which the second or even third empty orbital has the appropriate characteristics, namely that it lies at low energy and has $l > 0$ (Henry *et al.* 1969, Burke and Mitchell 1973). Although shape resonances have been found in the majority of molecules carefully studied at low energy, experimental verification of such effects in atoms

have been scarce until now. It is clear that this is a consequence of the limited degree to which excursions outside the noble gases have been made.

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