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PARAMAGNETIC SUSCEPTIBILITIES AND ELECTRONIC STRUCTURES
OF AQUEOUS CATIONS OF ELEMENTS 92 TO 95

Jerome J. Howland and Melvin Calvin

November 12, 1948

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Patent Advisor, Washington	52
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PARAMAGNETIC SUSCEPTIBILITIES AND ELECTRONIC STRUCTURES
OF AQUEOUS CATIONS OF ELEMENTS 92 TO 95

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ABSTRACT

Magnetic susceptibilities per gram atomic weight of elements 92 to 95 in most of their oxidation states were measured on 0.1 ml of solution which was 0.007 to 0.09 M in heavy element. The values obtained (all paramagnetic) in c.g.s. units $\times 10^6$ were: U(IV), 3690; Np(VI), 2060; Np(V), 4120; Np(IV), 4000; Pu(IV), 1610; Pu(III), 370; Am(III), 720.

The results could be interpreted only on the basis of electronic configurations $5f^n$, even though susceptibilities were generally lower than the theoretical values and lower than experimental values for corresponding lanthanide cations. The lower values should be expected as a result of the Stark effect produced by fields of anions and of water dipoles. Failure of the Russell-Saunders approximation to the coupling between electrons may account for some of the error in the theoretical calculations. Wider multiplet splitting in the actinides accounts for the fact that the susceptibilities of Pu(III) and Am(III) are many-fold lower than those of Sm(III) and Eu(III) respectively.

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Chemical and physical properties and theoretical calculations have indicated that elements of about atomic number 90 and higher constitute a series in which the 5f orbitals are being filled⁽¹⁾. These heavy elements have been called actinides⁽²⁾ in analogy to the name lanthanides for the rare earth elements. It was of interest to determine whether corresponding actinide and lanthanide aqueous cations have the same electronic configurations even though the actinide concept does not necessarily require that they be identical.

Electronic configurations can be determined unequivocally perhaps only from a study of optical spectra which may require years of work. If, however, an atom has its electrons in question (i.e., those in addition to the inert gas structure) in inner orbitals, then the electrons may be electrostatically shielded from neighboring atoms to the extent that the atomic axis of angular momentum is free to be oriented by an external magnetic field. Then the bulk magnetic susceptibility of a solution of such atoms can be deduced from quantum numbers of the ground state of the electronic configuration. Sometimes the converse, deduction of ground state quantum numbers from the susceptibility, will yield a unique answer. This was true for the lanthanide tripositive ions which have as outer configurations $4f^{1-14} 5s^2 5p^6$.

Susceptibilities of U(IV) and U(III) solutions have been reported by Lawrence⁽³⁾. Solid uranium compounds have been studied by numerous inves-

tigators⁽⁴⁾. At room temperature the susceptibility of U(IV) solutions and salts approximates the theoretical value derived from the spin angular momentum of two unpaired electrons with no contribution from orbital angular momentum. Since this type of calculation was successful in accounting for the susceptibility of the first row transition element ions which have partially filled 3d electron orbitals, the observed susceptibility of U(IV) was usually interpreted as evidence for a $5d^2$ electron configuration. This deduction completely ignored the fact that the observed susceptibilities of cations of heavier transition elements (those with partially filled 4d or 5d orbitals) are generally many-fold smaller than the "spin only" calculations. Hutchison and Elliott⁽⁵⁾ have interpreted their recent measurements on uranium (IV) salts to indicate a $5f^2$ structure.

Shortly after plutonium became available, Calvin⁽⁶⁾ measured the susceptibilities of dilute Pu(VI), Pu(V), Pu(IV), and Pu(III) solutions with the expectation that they might closely parallel those of Pr(III) through Sm(III) if the actinide ions also had f^n electronic configurations. The measurement of Pu(V) was very crude because of the instability of that state⁽⁷⁾. The results for the other three plutonium oxidation states were sufficiently accurate, but there was no close agreement with expectations of particular electronic structures. Some of the electrons had to be 5f, however. More actinide elements which exist in one or more oxidation states could be used in the present study. Since the alpha activity of the available isotope of curium would rapidly decompose the water of its aqueous solution, experiments with this material were not attempted.

Experimental

Magnetic susceptibility measurements were made on 0.1 ml samples which were of the order of 0.01 M in heavy element by use of a bifilar suspension

method developed from one described by Theorell⁽⁸⁾. A divided glass capillary was suspended as shown in Fig. 1. The actinide solution was in the left compartment; distilled water was in the right. The capillary moved a distance of the order of 0.1 cm when the current through the magnet coils was 40 amperes. (Field strength directly between the pole faces was about 17,000 Gauss.)

The horizontal force on the capillary is very nearly equal to the component of its weight in the direction of movement

$$F = w \cos(90^\circ - \theta) = w D/L \quad (1)$$

where w is the weight of the capillary, θ is the small angle through which the fibers rotate, and D is the horizontal displacement which was observed in a microscope equipped with a traveling cross hair. One scale division on the knob corresponded to a distance of 8×10^{-5} cm or to a force of 6×10^{-7} gram on a 0.5 gram capillary.

Each solution was measured several times at magnet coil currents of 20, 30, and 40 amps. in order to establish that susceptibilities were always independent of field strength. Table I gives average displacements for 40 amps. only.

The molar susceptibility, χ , of a substance equals I_m/H where I_m is the magnetic moment of a mole of the bulk material and H is the magnetic field strength. The total force acting on a long cylinder of cross section A and whose axis passes through an inhomogeneous field is

$$F = \chi \underline{M} A (H_2^2 - H_1^2)/2000 \quad (2)$$

where \underline{M} is the molar concentration and H_1 and H_2 are the field strengths on the ends.

Equation (2) would hold only for a perfectly cylindrical sample; it was used only for estimation of the field strength from the displacements of nickel chloride solutions. Susceptibilities of other substances were

calculated on the assumption that molar susceptibility was proportional to D/M if w , A , H_1 and H_2 are held constant. The displacement was measured for each actinide solution in the same compartment and at the same magnet coil current as was done for a standard nickel chloride solution. Correction for diamagnetism of the solvent and of the anions and for non-uniformity of the capillary was made by subtraction of their experimentally determined displacement. If the molar susceptibility of nickel chloride at 20°C is taken as 4436×10^{-6} c. g. s. units⁽⁹⁾, then

$$\chi = 4436 \times 10^{-6} D' \frac{M_{\text{NiCl}_2}}{M} / \left(\frac{M}{M} D'_{\text{NiCl}_2} \right) \quad (3)$$

where D' is the displacement after application of the correction.

The actinide solutions could be prepared with at least 99% of the element in the desired oxidation state by the proper choice of anion. The U(IV) solution was prepared by dissolution of weighed, distilled UCl_4 in oxygen-free hydrochloric acid solution. The last step in the preparation of the neptunium, plutonium, and americium solutions was dissolution of a hydroxide which had been precipitated with ammonium hydroxide. The plutonium concentrations were based on a weighing. The neptunium and americium solutions were assayed by measurement of the rate of alpha particle emission of a small aliquot. The specific activities in counts/min./mg. were taken as 7.90×10^5 for Np ⁽¹⁰⁾ and 3.36×10^9 for Am ⁽¹¹⁾ if a thin sample is mounted on platinum and a counter geometry of 50% is used (52.0% of the disintegrations are counted⁽¹²⁾). These values are said to be probably better than $\pm 5\%$. If better specific activities are reported at a later date, the magnetic susceptibilities should be corrected proportionately. The quantity of neptunium or plutonium which was not in the desired oxidation state was checked by measurement of the characteristic optical absorption maxima⁽¹³⁾ on a Beckman spectrophotometer.

The 0.03920 M nickel chloride solution which served as magnetic standard was prepared by dissolution of 0.2301 g. of nickel rod (Johnson Matthey and Co., 99.97% Ni) in 5 ml of refluxing 10 M HCl. After the solution had been diluted to 100.0 ml, the excess HCl concentration was found to be 0.360 M.

Although the measured displacements were reproducible to about one scale division, the uncertainty in the molar susceptibilities is about 2% or 30×10^{-6} units, whichever is larger, because of the inaccuracy in determination of actinide concentration and because of the presence in the solutions of an unknown amount of diamagnetic ammonium ion. The large negative displacements listed for some examples of solvent only in the rear compartment are due to non-uniformity of the glass capillary. These large subtractions do not increase the percent error if the gross displacement of the actinide solution is positive.

Results and Interpretation

Observed displacements and susceptibilities are given in Table I. In Fig. 2 experimental χ of the actinide cations are compared with simple theoretical χ_J for the lowest energy quantum states of the electronic configurations $5f^n$ and $5f^{n-1}6d$. The experimental χ follow the χ_J for configurations f^n to a significant degree, though not as closely as had been found for most of the lanthanide cations⁽¹⁴⁾. The susceptibility of Am(III) is much higher than the theoretical value of zero, but that is also true of the corresponding lanthanide ion, Eu(III). It will be discussed in a following section of this paper.

The ground states of the cations whose susceptibilities are plotted in Fig. 2 must be presumed to be those on which the theoretical curve B was based, i.e., $^2F_{5/2}$, 3H_4 , $^4I_{9/2}$, 5I_4 , $^6H_{5/2}$, and 7F_0 respectively as n is 1 through 6.

While the atomic quantum numbers of a state may be those which are expected for a definite configuration, f^n , the state can belong partly to another configuration of the same parity (i.e., "oddness" or "evenness"). This was shown theoretically by Condon⁽¹⁶⁾, and it accounts for a number of anomalies in spectra of complex atoms. Susceptibility measurements can show only that the ground state has certain L, S, and J quantum numbers; identification of the state with a configuration has become customary, but it is only an approximate concept.

To the extent that quantum states of complicated atoms can be attributed to a single electronic configuration, the known aqueous cations of uranium and higher elements must have as the outerpart of their ground configurations, $5f^n 6s^2 6p^6$ (n 5f electrons which are more or less inside the configuration for the inert gas element 86).

Approximate Nature of Theoretical Calculations

The simple theory of atomic structure as related to magnetic susceptibility is frequently inadequate. A few of the complications which may be important in the actinide cations should be mentioned. Quantum statistically the susceptibility is given by the relation⁽¹⁵⁾

$$\chi = \frac{I_m}{H} = \frac{N_0}{H} \frac{\sum \partial W_i / \partial H \exp(-W_i/kT)}{\sum \exp(-W_i/kT)} \quad (4)$$

where N_0 is Avogadro's number and the summations are over all energy levels, W_i . Usually it is sufficient to consider levels up to about 10 kT. Just how these levels depend on H is not always known. If the W_i are simply the Zeeman levels from an isolated (on the energy scale) J state of a free atom, their energies might be assumed to be

$$W_i - W_i^0 = Hg\beta M \quad (5)$$

where $\beta = eh/4\pi mc$ and $M = J, J - 1, \dots -J$. Then the first two terms of series approximations for each of the $2J + 1$ terms of the numerator and of

the $2J + 1$ terms of the denominator of equation (4) gives for the summation

$$\chi_J = N g^2 \beta^2 J(J + 1) / 3kT \quad (17) \quad (6)$$

If the orbital and spin angular momenta of the several electrons are coupled according to the Russell-Saunders scheme (LS coupling)

$$g = [3J(J + 1) + S(S + 1) - L(L + 1)] / 2J(J + 1) \quad (7)$$

Equations (6) and (7) were used to calculate the theoretical points of Fig. 2. The ground state of a configuration was assumed by application of Hund's rules. This calculation gives the "free atom" susceptibility. The limited agreement of theoretical and experimental susceptibilities indicates that each paramagnetic atom interacts with other surrounding atoms as well as with the external magnetic field. Since most of the surrounding atoms are diamagnetic, the interaction is probably electrostatic in nature.

Penny and Schlapp have attempted to calculate the effect of crystal electric fields on the magnetic susceptibility of iron group compounds and rare earth salts^(18,19). Qualitatively it is a Stark splitting which is of the order of kT whereas the Zeeman splitting is small compared to kT . Although the calculations are not perfected⁽²⁰⁾ it is clear that the summation (4) can be much smaller than its evaluation by equation (6). For solutions of paramagnetic ions the same picture can be used qualitatively to describe the effect of electric fields of neighbor anions and water dipoles. The magnetic susceptibility of an actinide cation should be expected to be lower than its corresponding lanthanide ion since the less perfectly shielded f electrons of the former are more vulnerable to electric fields. Complex ion formation will not in itself affect the susceptibility unless the f electrons are involved directly in chemical bond formation. The greater proximity of a negative radical in a complex could, however, accentuate the Stark effect.

Since the example of Np(VI) is essentially a one-electron case, the complications to be mentioned later are not possible, and the lowering of the susceptibility from the theoretical χ_J must be due to the Stark splitting. It is logical to conclude that the Stark effect will be the most important single complication tending to lower the susceptibility of all of the actinide cations.

Ground quantum states of the $4f^n$ configurations were deduced by Hund⁽¹⁴⁾ from rules which are correct for relatively light elements in which it is a good approximation to derive atomic states from the states of individual electrons by means of the LS or Russell-Saunders coupling model. For elements of very high atomic number the LS coupling approximation does not give the order in which the various J states fall on the energy scale. It has been observed in the spectra of gaseous uranium^(21,22) that the multiplet splitting is wider than the separation of the centers of gravity of LS states--the opposite of the situation for good LS coupling. The experimental g's are fairly close to g_{LS} , however. The spectra also show that the lowest levels of neutral uranium belong to the configuration $5f^3 6d 7s^2$.

It is interesting to see what happens to the J levels as the spin-orbital interaction approaches the limiting case of jj coupling. Column 2 of Table II lists some J states which should lie very low in the configurations $5f^{1-6}$. The state listed first lies deepest according to Hund's rules for LS coupling. For $5f^2$, $5f^3$, and $5f^4$ the state listed second was selected because it belongs both to the next LS group and to the lowest group of jj coupling states. In the case of $5f^6$ the two lowest J states of 7F were selected because the spacing between $J = 0$ and $J = 1$ would be only $1/21$ of the total multiplet splitting according to the Landé interval rule. The ${}^6H_{5/2}$ state of $5f^5$ becomes more isolated as jj coupling is approached. The relative importance of the probably second lowest states is not known at

the present time. Their theoretical χ_J 's as calculated according to equation (6) should be averaged with the χ_J 's of the lowest state according to the statistical factor $(2J + 1) \exp(-W_J/kT)$.

The two theoretical χ_J 's for each ground state of the $5f^n$ configurations diverge as n increases from 2 to 5. Since the two χ_J 's differ directly as g_{LS}^2 and g_{jj}^2 differ, the experimental χ of Pu(III), the example of $5f^5 \ ^6H_{5/2}$, shows unambiguously that g is much nearer to g_{LS} than to g_{jj} . This is consistent with previously mentioned results for the gaseous uranium atom.

Equation (6) is sometimes not adequate even for an atom which has none of the complications mentioned above. For each low energy J state of Sm(III) and Eu(III) it was necessary⁽²³⁾ to use a more complete equation for χ_J which had been derived by Van Vleck⁽¹⁵⁾ from considerations equivalent to taking the Zeeman splitting to the second order terms in H

$$\chi_J = \frac{Ng^2\beta^2 J(J+1)}{3kT} + \frac{N\beta^2}{6(2J+1)} \left\{ \frac{f(J)}{h\nu(J-1;J)} + \frac{f(J+1)}{h\nu(J+1;J)} \right\} \quad (8)$$

$$\text{where } f(J) = \frac{[(S+L+1)^2 - J^2] [J^2 - (S-L)^2]}{J}$$

For sufficiently large $h\nu$'s equation (8) reduces to equation (6) except that when S and L are large and J is small, the second term is extremely large.

In the examples Pu(III) and Am(III) the susceptibilities are greater than theoretical χ_J 's for $^6H_{5/2}$ and 7F_0 (Note the crossing of curves A and B in Fig. 2.) as obtained with equation (6). The second term of equation (8) must be significant in low lying states of both of these cations. Also the 7F_1 state must lie low enough in Am(III) to be well populated, though much less so than in Eu(III) where the multiplet splitting is narrower. Thus the same discrepancies with simplest theory that were important in Sm(III) and in Eu(III) can be detected in Pu(III) and Am(III), but they are

smaller and are nearly hidden by other complications which are common to all of the actinide ions. The analogy is shown clearly in Fig. 3 where experimental susceptibilities of lanthanide and actinide cations are compared directly.

Acknowledgement

The neptunium and americium were available for this study only through the work of T. J. LaChapelle, L. B. Magnusson, L. B. Asprey, and B. B. Cunningham, who had isolated these materials in very pure form.

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Table I
Susceptibilities at 20°C from Average Displacements
for 40 Ampere Magnet Coil Current

Material in Rear Compartment.	Displacements		$\chi \times 10^6$ cgs emu
	D	D'	
Capillary 2B, 0.577 g			
0.36 M HCl	-	240	
0.0392 M NiCl ₂ in 0.36 M HCl	+1060	1300	4436 ^a
0.50 M HCl	-	270	
0.0492 M U(IV) in 0.5 M Cl ⁻	+1088	1358	3690 ^b
0.20 M HCl	-	240	
0.0140 M Np(V) in 0.2 M Cl ⁻	+ 191	431	4120 ^b
0.20 M H ₂ SO ₄	-	237	
0.0160 M Np(IV) in 0.2 M HSO ₄ ⁻	+ 241	478	4000
0.50 M H ₂ SO ₄	-	283	
0.0924 M Np(VI) in 0.5 M HSO ₄ ⁻	+1139	1422	2060
Capillary 9A, 0.420 g			
0.36 M HCl	-	30	
0.0392 M NiCl ₂ in 0.36 M HCl	+ 991	1021	4436 ^a
0.50 M HCl	-	30	
0.0600 M Pu(III) in 0.5 M Cl ⁻	+ 99	129	370
0.50 M H ₂ SO ₄	-	89	
0.0507 M Pu(IV) in 0.5 M HSO ₄ ⁻	+ 389	478	1610
Capillary 11A, 0.518 g			
0.36 M HCl	+ 131		
0.0392 M NiCl ₂ in 0.36 M HCl	+ 999	868	4436 ^a
0.50 M HNO ₃	+ 156		
0.0908 M Am(III) in 0.5 M NO ₃ ⁻	+ 483	327	720

^aMagnetic standard, ref. 9.

^bPu(VI), which has the same number of electrons as U(IV) and Np(V), has a $\chi \times 10^6$ of 3540, ref. 6.

Table II
Theoretical Susceptibilities of Possible Low Energy
States of Actinide Ions

Assumed Electronic Configuration	Possible Low Energy States	Theoretical χ_J 's LS Coupling 20°C	Theoretical χ_J 's jj Coupling 20°C
$5f^1$	$^2F_{5/2}$	2730	2730
$5f^2$	$^3H_4; ^3F_2$	5420; 1130	6210; 1870
$5f^3$	$^4I_{9/2}; ^4G_{5/2}$	5540; 1210	7680; 2730
$5f^4$	$^5I_4; ^5G_2$	3040; 280	6210; 1870
$5f^5$	$^6H_{5/2}$	300	2730
$5f^6$	$^7F_0; ^7F_1$	0; 1900	0; 1900
$5f6d$	$^3H_4; ^3G_3$	5420; 2860	
$5f^2 6d$	$^4K_{11/2}; ^4I_{9/2}$	8950; 5540	
$5f^3 6d$	$^5L_6; ^5K_5$	9070; 5670	
$5f^4 6d$	$^6L_{11/2}; ^6K_{9/2}$	5730; 3130	
$5f^5 6d$	$^7K_4; ^7I_3$	1350; 320	

Fig. 1.--Apparatus for measurement of magnetic susceptibility: A, glass fibers, 0.005 x 140 cm; B, glass capillary, 0.2 x 16 cm; C, magnet pole faces, 2.5 cm diam.; D, mirror and microscope; E, magnet pole pieces, 15 cm diam.

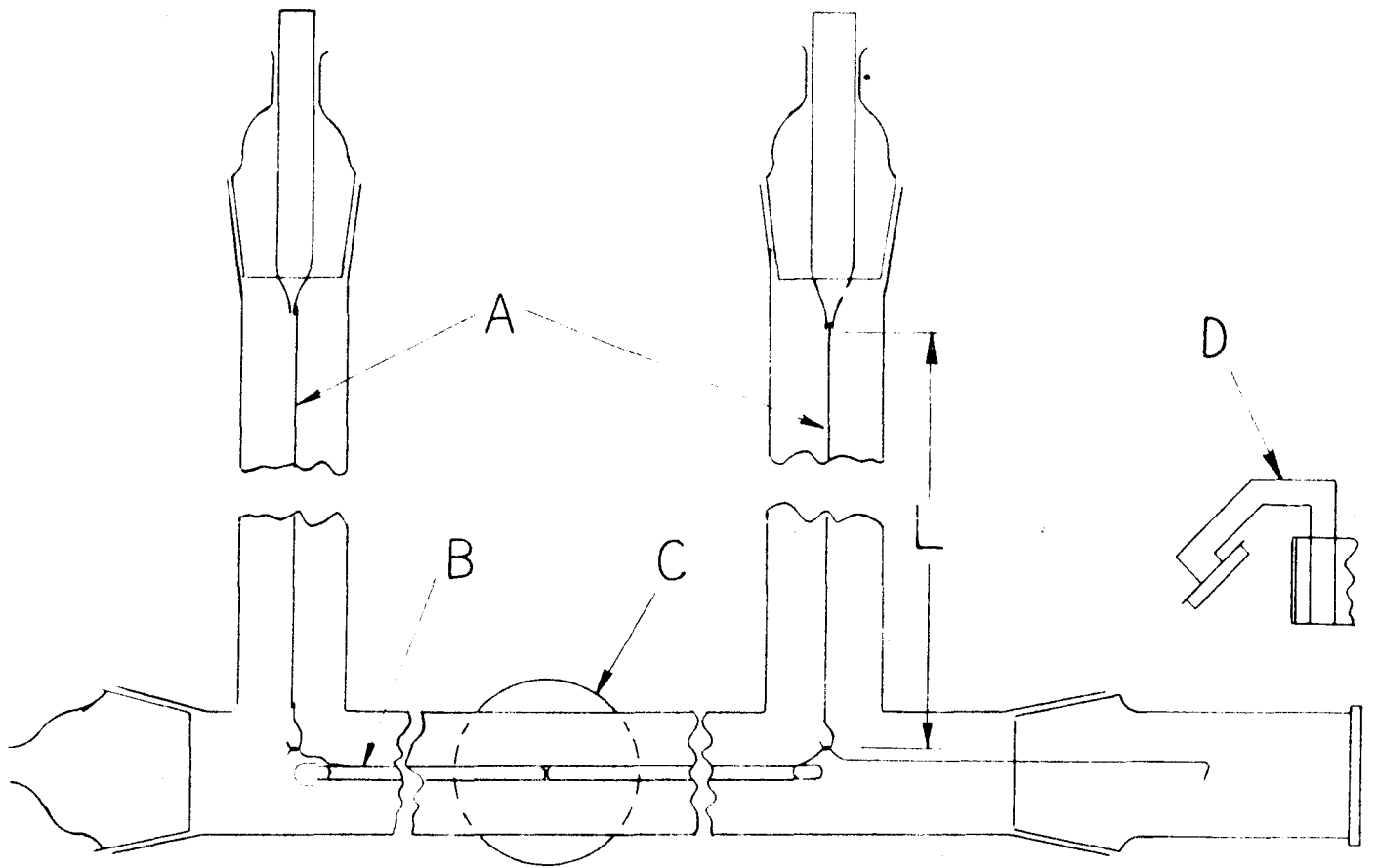
Fig. 2.--Comparison of magnetic susceptibilities of actinide cations with theoretical susceptibilities: A, experimental; B, theoretical for ground states of $5f^n$; C, theoretical for ground states of $5f^{n-1}6d$.

Fig. 3.--Comparison of experimental magnetic susceptibilities of lanthanide and actinide cations at 20°C. The values for the lanthanides were calculated from some of the "effective magnetic moments" compiled by Yost, Russell and Garner⁽²⁴⁾. The point for 61(III) is the theoretical calculation by Van Vleck and Frank⁽²³⁾.

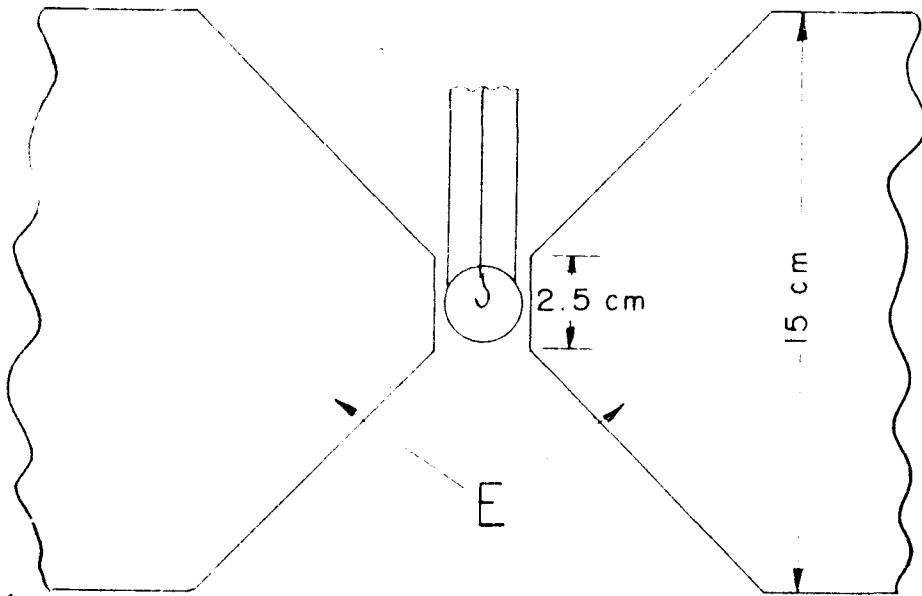
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17. The small contribution to susceptibility from the magnetic moment induced by the applied field was not added to the theoretical χ_J 's. It would be only about -50×10^{-6} for monatomic ions represented by Pu^{+3} and Am^{+3} and possibly as much as $+50 \times 10^{-6}$ for molecular types such as NpO_2^+ and NpO_2^{++} .
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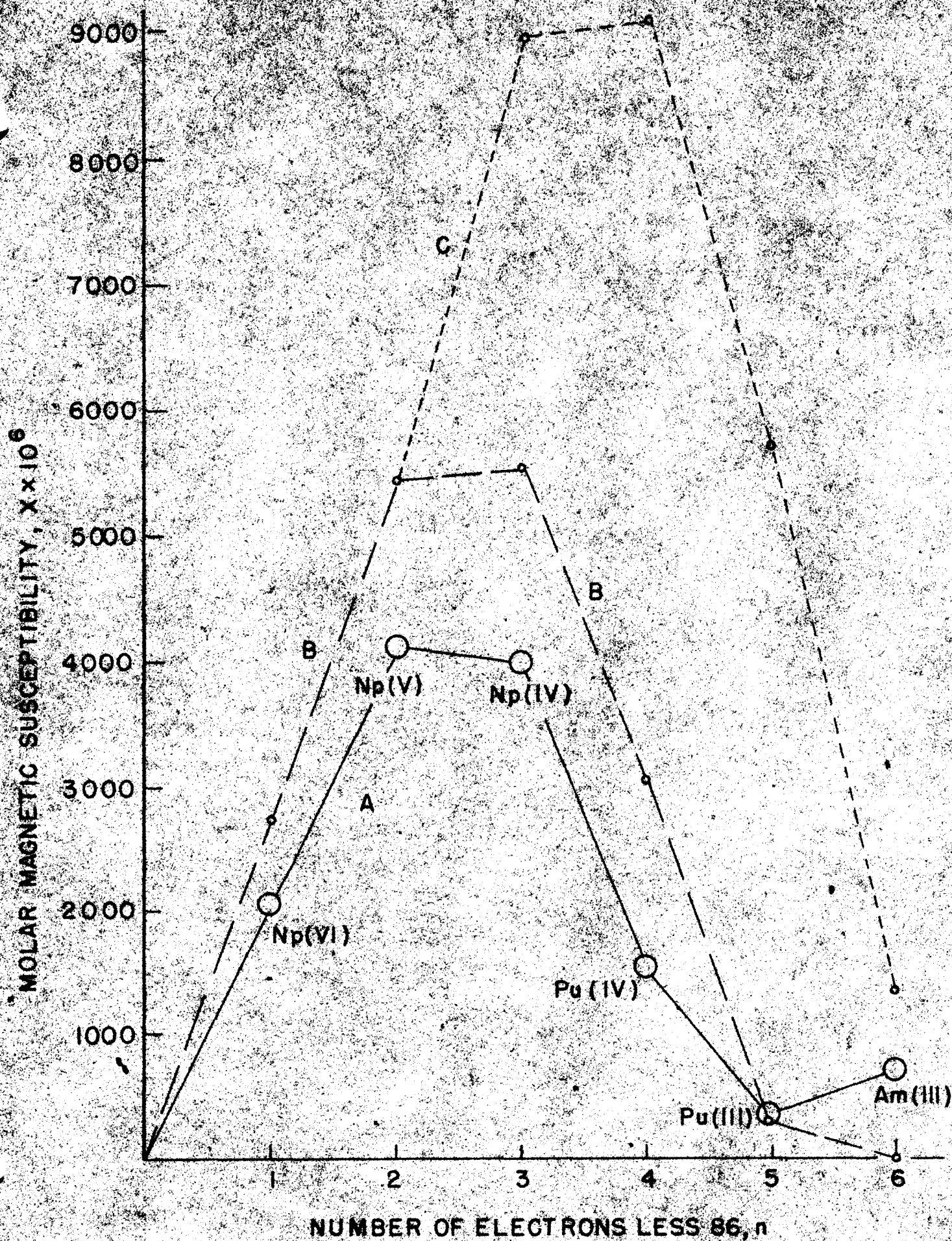


FIG 2

