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Autor(en): **Enz, Charles P.**

Objektyp: **Article**

Zeitschrift: **Helvetica Physica Acta**

Band (Jahr): **56 (1983)**

Heft 5

PDF erstellt am: **22.07.2024**

Persistenter Link: <https://doi.org/10.5169/seals-115429>

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Heisenberg's applications of quantum mechanics (1926–33) or the settling of the new land*)

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(10. I. 1983)

1. Introduction

The discovery of quantum mechanics between 1925 and 1926 was almost simultaneously followed by the 'setting out for the new land' ('Der Aufbruch in das neue Land', title of Chapter 6 of Ref. 1) to explore the new theory with respect to its physical interpretation and its formal structure. Heisenberg played an important part in both these stages.

In his exploratory role Heisenberg mainly contributed as co-author of the '3-man paper' ('3-Männerarbeit', see Ref. 2, letter [102], and Ref. 3, p. 95) with Born and Jordan [4] but also as partner of Pauli in an exchange of letters on the development of a formalism of action and angle variables (see Ref. 2, letters [117] and [118], and Ref. 3, Section V.4). This endeavour was never published because the problem had just been solved in a most elegant way by Dirac [5].

In a third stage devoted to the 'settling of the new land' quantum mechanics was applied to the physical problems left unexplained by the old quantum theory. Here again Heisenberg made important contributions which are described in this paper.

Prominent among the unsolved physical problems was the anomalous Zeeman effect with which Heisenberg had struggled since the beginning of his scientific career and whose solution he got at last in 1926 (Section 2). A more difficult problem, because it involves two electrons, was the singlet-triplet splitting in the spectrum of the Helium atom which Heisenberg solved in the same year (Section 3). The third application concerned an old many-electron problem, namely the explanation of ferromagnetism; it was settled by Heisenberg in 1928 (Section 4).

After these major breakthroughs Heisenberg's interest turned to quantum field theory, cosmic radiation and nuclear structure. Only occasionally did he come back to particular problems suggested by experiments such as electrons and holes in atoms and in solids (Section 5) and incoherent X-ray scattering (Section 6).

*) Extended version of the Introduction to Group 4 of the Collected Works of Werner Heisenberg.

2. Action-angle variables and the anomalous Zeeman effect

The action-angle variables proved their usefulness in Heisenberg's application of the new theory to the anomalous Zeeman effect (see Ref. 3, Section V.5). In this work which he published with Jordan [6] the spectroscopic effect of the Zeeman energy H_1 , of the spin-orbit coupling H_2 and of Sommerfeld's relativistic correction H_3 [7] was calculated in first order perturbation theory. H_2 and H_3 contained the orbital averages $\langle r^{-3} \rangle$ and $\langle (W_0 + Ze^2/r)^2 \rangle$, respectively, and the only way at that time to calculate $\langle r^{-2} \rangle$ was by making use of the action-angle variables. The calculation of $\langle r^{-3} \rangle$, on the other hand, could have been based on Pauli's brilliant matrix-mechanical solution of the hydrogen atom [8]. But the authors preferred to rely upon the simpler, although quite unphysical, 2-dimensional hydrogen model investigated by Dirac at the end of Ref. 5. Of course, Schrödinger's theory would have offered a more systematic method to calculate these averages (see Ref. 9, p. 103). But the date of submission of Schrödinger's third note [10] which proves the equivalence of his theory with matrix mechanics (see also Pauli's letter to Jordan of April 12th, 1926, Ref. 2, letter [131]) was March 18th, 1926 while Heisenberg and Jordan submitted their paper on March 16th, 1926. In addition, however, the techniques described in Ref. 9 had first to be worked out.

Already in a letter to Pauli of December 24th, 1925 (Ref. 2, letter [112]) Heisenberg notes that the problem of the anomalous Zeeman effect could easily be solved if the averages $\langle r^{-2} \rangle$ and $\langle r^{-3} \rangle$ were known. The crucial condition at that time, however, was the acceptance of Goudsmit and Uhlenbeck's model of the electron spin [11] which actually had already been proposed by A. H. Compton in 1921 based on the results of his investigations of ferromagnetic substances with X-rays [12]. But the work on this problem was hampered by Pauli's well-known opposition to the spin idea. As late as March 8th, 1926 Pauli explains his objections in a letter to Kramers (Ref. 2, letter [125]) but 4 days later he surrenders in a letter to Bohr (Ref. 2, letter [127]). This opened the road for the exploration of all the other problems in which the electron spin played a crucial role (see the following sections).

3. Exchange and the Helium atom

The problem of the singlet-triplet splitting in the spectra of two-electron systems belonged to a program of urgent research problems agreed upon between Bohr and Pauli in a conversation they had in Berlin in December 1925 (see Ref. 2, p. 311 and the references given there). Early in May of the following year Heisenberg informs Pauli that he has found 'a rather decisive argument that your prohibition of the equivalent orbitals is related to the singlet-triplet spacing' (see the postcard [132] in Ref. 2). This recognition of the role of Pauli's exclusion principle was the key for Heisenberg's success with the problems of more than one electron, most importantly for his two famous papers on the Helium spectrum [13] (see Ref. 3, Section V.6) and on ferromagnetism [14] (see Section 4). Both these problems arose from the realization that the magnetic (spin-spin) interaction energies were far too small to serve as an explanation; in both cases the subtle combination of Coulomb repulsion and Pauli principle leading to exchange was the answer discovered by Heisenberg.

In a first paper submitted June 11th, 1926, Heisenberg 'tries to give the foundation for the quantum mechanical treatment of the many-body problem' [15]. This paper already gives the qualitative solution of the problem of singlet-triplet splitting by introducing the notion of quantum mechanical resonance due to exchange degeneracy of non-equivalent states. But its historical significance lies in the fact that it stresses for the first time the implication for identical particles (electrons) of the exclusion principle (this 'housing office for equivalent electrons' as Pauli calls his principle in a letter to Wentzel, Ref. 2, p. 322), namely the determinantal form of the n -particle states and the reduction by a factor $n!$ of the statistical weight of these states [16]. Heisenberg also tries to show by induction from n to $n + 1$ that a system which is described by a determinantal wave function 'is the only one which contains no equivalent states of the partial systems' [15] but he does not quite succeed [17].

The main paper on two-electron systems [13] was submitted on July 24th, 1926, only $1\frac{1}{2}$ months after the first [15]. Heisenberg took this investigation on the Helium atom as an opportunity to acquaint himself with Schrödinger's new methods (see Ref. 1, p. 104; English translation p. 72); indeed, he does a detailed, even numerical calculation. 'Why should one not once in a while ride a steam roller' he writes to Pauli in the letter announcing this paper (Ref. 2, p. 328). The main result is that the energy of the Para (antisymmetric) state is higher than that of the Ortho (symmetric) state by twice the exchange integral $H^1(vw, vw)$ while the perturbation of the middle between the Ortho- and Para-levels is given by the Coulomb integral $H^1(vw, vw)$. In a first part, numerical values of these integrals are given for $Z = 2$ (He) and $Z = 3$ (Li^+), v being the $n = 1$ S-state and w the P- and D-states with $n = 2$ and 3. In spite of having taken into account shielding and the Rydberg and polarization corrections Heisenberg is not satisfied with the attained accuracy (see his concluding remarks). In fact, as can be seen from Ref. 9 this problem has seen a long sequel of approximation methods.

In a second part [13] the effect of the electron spin ('Elektronenmagnet') through spin-orbit coupling (H_1) and spin-spin dipolar coupling (H_2) is analyzed in a discussion which on p. 512 is somewhat heavy: Parallel spins (states a and d) have 'equivalent partial systems' ($m_1 = m_2$) and hence have no resonance degeneracy while antiparallel spins (states b and c) do since they are non-equivalent ($m_1 \neq m_2$). But a and d combine with the symmetric part $b + c$ of the 'resonance problem' b, c to form the triplet $a, b + c, d$ (not a, b, c as stated, and the singlet not c but $b - c$). But this discussion is interesting because it clearly shows the correspondence between the ortho-para separation of the orbital states and the triplet-singlet separation of the spin states.

While for two electrons the consequences of the above correspondence are still trivial this ceases to be so for 3 or more particles, which is the subject of the third paper of this trilogy [18]. The reason is that for 3 or more particles parallel spins are unavoidable and prevent totally antisymmetric spin states. Most significantly, Heisenberg notes (p. 253 of Ref. 18) that 'for [spin] values $s > \frac{1}{2}$ such a consideration would not be realizable'. With this in mind the correspondence noted by Heisenberg may be considered as a precursor of Dirac's fundamental theorem [19] which was the basis for his vector model [19] (see Section 4). But this third paper is noteworthy mainly because it is Heisenberg's first experience with group theory and together with the simultaneous first paper by Wigner on the same subject [20] inaugurates the year of the group theory fad in *Zeitschrift für Physik* to which we will come back in the next section.

4. Exchange and ferromagnetism

By introducing his idea of a molecular field [21] into Langevin's theory of the magnetism of classical dipoles, Weiss was able to account for most of the phenomenology of ferromagnets [22]. However, he himself realized that this field could not be due to magnetic interactions. On the other hand, the absence of electric dipole moments in the groundstate of atoms [23] suggests, as Heisenberg observes in the introduction to his fundamental paper [14] 'that the electric interactions between two atoms should be proportional to the square of the cosine of their mutual angle rather than proportional to the cosine, contrary to the assumptions of Weiss' theory'.

Heisenberg's paper [14] which was submitted on May 20th, 1928 brought the break-through sought during 20 years. He had the lucky inspiration to see that, as he says in §1, 'the empirical results with ferromagnetism place us into a very similar situation as the one we had encountered once earlier with the spectrum of the Helium atom'. Indeed, the exchange interaction was once again the key to the puzzle, an idea Heisenberg had expressed already in a letter to Pauli of November 1926 (see Ref. 1, p. 353). The problem stated in §2 is to find the distribution of energy levels ('Termwerte') of $2n$ localized electrons (1 valence electron per atom), to first order in the coupling by the exchange integral J_0 between each electron and its z nearest neighbours. Couplings among more distant electrons are negligible because of the exponential decrease of the exchange integrals.

In a second step the partition function is calculated. Here Heisenberg makes, as he admits, the 'somewhat arbitrary' assumption of a Gaussian distribution around the mean energy E_σ with variance ΔE_σ^2 . To calculate E_σ and ΔE_σ^2 Heisenberg follows the group theory fashion that blossomed in *Zeitschrift für Physik* all through 1927 and which, as we saw in Section 3, had been inaugurated by himself and by Wigner. Today both, terminology and technique developed in this literature are unfamiliar to most physicists. As to terminology the reason of the difficulty is that during this hectic activity of 1927 the older mathematics literature apparently was overlooked (see footnote 16 to Chapter V, p. 408 of Ref. 24) while the introduction of the spin-Hamiltonian concept by Dirac [19] made the calculation of group characters unnecessary (see below).

The 'Partitio' in Eqs (4) and (3) of Heisenberg's paper [14] are, respectively, a Young pattern and its dual (see Chapter V, § 13 of Ref. 24), restricted to 2-dimensional representations (spin $\frac{1}{2}$); the rows of a Young pattern characterize the cycles of a permutation of the $2n$ electrons and the dual has rows and columns interchanged. Permutations of the electron's spin independent of its orbit means of course that spin-orbit interactions are neglected in this approach. As to the calculation of the characters in Eq. (13) of Ref. 14 they follow easily, e.g., from Eq. (13.15), p. 138, of Ref. 25.

This last quoted formula, however, as all calculations in Heisenberg's paper, is valid only for spin $\frac{1}{2}$ (1 valence electron per atom). In his contribution to Sommerfeld's 60th birthday [26] Heisenberg has generalized his theory to y valence electrons per atom which are assumed to obey Hund's rule ('ein Elektron in einer Quantenzelle'), i.e. to give rise to an atomic spin of $y/2$. However, as van Vleck has remarked in Footnote 16 on p. 329 of his famous book [27] this elaborate calculation of ΔE_σ^2 is wrong since Heisenberg has omitted the contribution of two independent transpositions T_{12} and T'_{12} in Eqs (8) and (9) of Ref. 26.

The motivation for this generalization to $y > 1$ was that in his first paper [14] Heisenberg did not get the Langevin function [22] $\coth x - 1/x$ but $\tanh x$. The latter followed from the logarithmic derivative of the partition function S in Eq. (22a) of Ref. 14. In his second paper [26] the number of terms $f(m)$ in the expression (16) for S is defined through the identity

$$S \propto \sum_{m=-ny}^{+ny} f(m)z^m = \left(\sum_{k=-y/2}^{+y/2} z^k \right)^{2n} \quad (\text{I})$$

For $y = 1$ this identity reduces to Eq. (22a) of the first paper [14] and then is nothing but the coefficient of x^m for $\lambda_1 = \lambda_2 = \dots = 1$ ($\sum \lambda_i = 2n$) in Eq. (13.14) of Ref. 25. This is a remarkably clever way of transforming the partition function into a sum over the magnetic quantum number m . Note that for $f(m) = 1$ the logarithmic derivative of equation (I) above leads to the Brillouin function [22] for angular momentum ny . But in spite of the mentioned deviation from the Langevin function Heisenberg was able to reproduce the main features of Weiss' theory and thus give an explanation of ferromagnetism in terms of the exchange integral J_0 .

From the point of view of perturbation theory the Gaussian distribution introduced by Heisenberg in Ref. 14 is not convincing. Indeed, after integration over it the partition function S , Eq. (20), has the familiar canonical form with weight f_σ , except for the term coming from the variance ΔE_σ^2 which is second order in J_0 while perturbation theory had been carried only to first order in J_0 . In fact, Pauli in his exhaustive report to the Solvay Council on magnetism [28] of October 1930 starts precisely at this point, namely with Eq. (20) of Ref. 14 and leaving out the questionable term. But instead of transforming to a sum over the magnetic quantum number m Pauli determines the most probable spin value \bar{s} and shows that, to relative order n , $\bar{m} = \bar{s}$. This immediately yields the magnetization and Heisenberg's result for spin $\frac{1}{2}$, Eq. (24) of Ref. 14, except for the terms of second order in $\beta = zJ_0/kT$ (see Eqs (64)–(66) of Ref. 28). This critique also shows that Heisenberg's condition for ferromagnetism, $z \geq 8$ [14], is not to be taken seriously although, as he points out, it is satisfied for Fe, Co, Ni. Heisenberg has reported his work on ferromagnetism in a series of letters to Pauli predating the submission of the manuscript (Ref. 2, letters [192]–[196] and [198]). In the first of these letters the partition function is the same as Pauli's, and only in the second letter Heisenberg worries about the level distribution around the mean energy E_σ with variance ΔE_σ^2 .

Van Vleck in his mentioned book [27] follows Heisenberg's statistical evaluation more closely, except for using the method of steepest descent. But his presentation of Heisenberg's theory is of considerable historical importance because, stimulated by Dirac [19], he was the first to use consistently the spin Hamiltonian (Eq. (16) of Ref. 27)

$$\mathcal{H}_{\text{spin}} = - \sum_{\langle i, j \rangle} J_{ij} \vec{S}_i \cdot \vec{S}_j \quad (\text{II})$$

where $\langle i, j \rangle$ are pairs of nearest neighbours (in the above notation $J_{ij} = J_0$). Most importantly, in §78 van Vleck gives a proof of Eqs (14) and (17) of Ref. 14 'derived by Heisenberg with the rather involved machinery of group theory' showing 'that Dirac's kinematical interpretation of the exchange effect ... frees us

from the need of using this' [27]. The frustration from the 'group pest' (see Ref. 24, preface to the second edition) combined with the popularity of van Vleck's book precipitated the new look of equation (II) above. But it is still surprising how this expression could become called 'Heisenberg Hamiltonian' or 'Heisenberg model' [22] since it surely is vain to look for it in Refs 14 and 26. As far as I can trace it (see also Ref. 29, p. 16) this name was also promoted by van Vleck who uses 'Heisenberg model' in the title of the third section of his review of 1945 [30].

Although the spin Hamiltonian became quite popular, its justification remained controversial. This must have been due, at least in part, to the fact that in higher than first order $\mathcal{H}_{\text{spin}}$ is not unique. But Herring in his thorough review assures us that 'doubts that have been expressed in the literature about this form are unfounded' [29].

Even more controversial is the history of the sign of the exchange integral J_0 : Heisenberg who was fully aware of the difficulty of this question came to the conclusion that 'for sufficiently high principal quantum numbers J_0 is certainly positive' ($n \geq 3$) [14]. Herring, however, observes that for interatomic spacings large enough for the spin Hamiltonian (II) to be valid J_{ij} is almost certainly negative (antiferromagnetic) whenever i and j are nearest neighbour atoms of the same kind (see Ref. 29, p. 6 and p. 59). Magnetism in fact has turned out to be an even more complex phenomenon than Heisenberg had anticipated on p. 116 of Ref. 26. New exchange mechanisms have turned out to be important: superexchange [31], indirect exchange and itinerant exchange [22], the latter being responsible for the ferromagnetism of transition metals such as iron and nickel [32]. How complex the problem is for these two most common ferromagnets may be appreciated from a recent numerical analysis [33].

In Ref. 26 Heisenberg made the interesting prediction that the elastic constants of a ferromagnet should change when traversing the Curie temperature. There is indeed experimental evidence in Ni [34] and in some Fe-Ni alloys [35] but the theoretical understanding is still limited [36]. In a later paper [37] Heisenberg made a similar prediction concerning the stress-strain relation of iron: the curve should show a discontinuous change of slope at the strain corresponding to the (spontaneous) saturation magnetostriction [38]. Here no experimental results are known to me.

This third paper on magnetism by Heisenberg [37] is purely phenomenological and is based on the much weaker magnetic (as opposed to the electric exchange) interaction between the atoms and on the ensuing formation of domains [21, 22]. The walls separating these domains are the subject of the work in progress by Bloch [39] mentioned in Ref. 37. In a short review which slightly predates this third paper Heisenberg describes the actual knowledge of ferromagnetism to an industrial audience [40].

5. Electrons and holes

Heisenberg's work after his journey to the United States in 1929 (see Ref. 2, pp. 488, 515) showed less urgency than in the period before. One of the applications of quantum mechanics and, in particular of the exclusion principle, concerned the symmetry between holes and electrons in a filled atomic shell or in

a filled energy band of a crystal [41]. In this problem Heisenberg made use of the technique of second quantization which was familiar to him through his work with Pauli on quantum field theory done prior to his departure to America [42]. The second-quantized Hamiltonian expressed in terms of holes, Eq. (15) of Ref. 41, contains additional 1-particle operators arising from commutations of 2-particle operators. For an N -electron shell and n electrons this leads to the wave equation (17) or (37) of Ref. 41 for the $N-n$ holes which is nothing else than the Hartree-Fock approximation (see, e.g., Eq. (3.11) of Ref. 43) and which contains the energy A , Eq. (16) of Ref. 41, of the closed shell in the same approximation (see Eq. (3.8) of Ref. 43). Heisenberg seems not to have been aware of this connection with the work of Hartree [44] and of Fock [45]. The Hartree term $C(r_1)$ is just the Coulomb potential while the Fock term $D(r_1)$ is the non-local exchange potential. For a given N and neglecting spin interactions A just leads to a shift of the spectrum of n holes relative to that of n electrons. On the other hand, if one approximates the effect of the potentials $C(r_1)$ and $D(r_1)$ by an effective nuclear charge this leads to a factor of order one by which the n -hole spectrum is multiplied relative to the n -electron spectrum. A numerical comparison between the spectra of Ti and Ni confirms this conclusion.

In a second part [41] the electron-hole symmetry is applied to a metal with 'anomalous' Hall effect (see Ref. 22, p. 236). In the simplest case such a metal or a semiconductor has an almost filled conduction band in which the electrical current is carried by holes. By introducing a periodic potential eV_0 the Coulomb interaction between conduction electrons is taken care of so that there is no need for 2-particle operators in this case. Using a simple form of energy band, Eq. (44) of Ref. 41, and going over from Bloch functions, Eq. (43), to Wannier functions, Eq. (45), (see Ref. 22, p. 187) Heisenberg shows that for holes the Hall coefficient has opposite sign to that for electrons.

6. Incoherent X-ray scattering

It was Heisenberg's colleague in Leipzig, Peter Debye, who suggested that the Thomas-Fermi approximation used for the coherent i.e., phase dependent, part of X-ray scattering should be generalizable to the incoherent part. Heisenberg came up with an elegant and simple solution [46] entirely in the spirit of Thomas and Fermi (see, e.g., Ref. 23, p. 271). The latter amounts to treating the electrons in the atom as free except for the condition of binding, namely that the total energy in each point \vec{r} be negative,

$$\frac{\vec{p}^2}{2m} - e\phi(\vec{r}) < 0, \quad (\text{III})$$

$\phi(\vec{r})$ being the Thomas-Fermi potential. Free electrons means, in particular, that all orbital states are doubly occupied, corresponding to both spin directions, except perhaps for the highest one. Obviously, this is not a realistic approximation since it is the exact opposite of Hund's rule which is the consequence of the Coulomb repulsion. But here it is consistent. With the double occupancy use can be made of the closure of the spin states which leads to the $\delta_{\sigma\sigma}$, in Eq. (15) of Ref. 46. At this point the generalized Thomas-Fermi hypothesis (17) appears natural, and the result is straightforward.

A subsequent note by Bewilogua [47] gave a numerical table of Heisenberg's formula; and in a later short review Heisenberg came back to the definition of coherent and incoherent radiation [48]. This publication, unfortunately, is hampered by ambiguous notation since in Eqs (13) and (14) of Ref. 48 X_n stands both for a coordinate of the n th nucleus and for a wavefunction in state n . Although incoherent scattering has remained a side-line of X-ray research it is interesting to note that Heisenberg's formula has survived in the literature [49].

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