$$\operatorname{Im}\Pi_{pol}=\pi\sum_{s}\sum_{e'}\int\frac{d\mathbf{k}'}{2\pi}|F_s(\mathbf{ke}|\mathbf{k}',\mathbf{e}')|^2|q_s(\mathbf{k}-\mathbf{k}')|^2\delta(\omega-\omega'-\omega_s), \tag{24}$$

where F_s is the constant of the interaction of the light with the phonons, and **k** and **e** are the wave vector and polarization vector of the photon. Since the photon wave vectors **k** and **k**′ are very small, it suffices to calculate q_s at $k=0$. This can be easily done by using (6) and (7). The result is

$$q_s(k=0)=\frac{2^{7/2}i\pi\alpha}{1-\nu_s^2}\sum_{\mathbf{k}'}\frac{q_s(\mathbf{k}')}{k'}\sum_{n>0}\frac{(\psi_0 r_x\psi_n)(\psi_n e^{i\mathbf{kr}}\psi_0)}{E_0-E_n}=\frac{(2^{5/2}\pi\alpha)^{1/2}}{1-\nu_s^2}x_s, \tag{25}$$

where x_s is the coordinate matrix element given by (22) for the transition with excitation of the state q_s.

Since $x_s\sim\alpha^{-2}$, the ratio of the Raman-scattering intensity per polaron to the scattering intensity per crystal unit cell is of the order of

$$(\alpha^2 m\omega_0 a^2)^{-3/2}\gg 1, \tag{26}$$

where a^3 is the volume of the unit cell. The inequality sign accounts for the fact that we are working within the framework of the continual model polaron model, for which the applicability criterion is in fact (26). Thus the cross section for Raman scattering with excitation bound states of phonons is gigantic, as is the cross section for the absorption of light.

The author thanks E. I. Rashba for a number of useful discussions.

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Translated by J. G. Adashko

Calculation of the energy of the molecular phase of solid hydrogen allowing for three-particle interactions

S. I. Anisimov and Yu. V. Petrov

L. D. Landau Institute of Theoretical Physics, USSR Academy of Sciences, Moscow

(Submitted 15 September 1977)
Zh. Eksp. Teor. Fiz. **74**, 778–785 (February 1978)

A calculation is made of the static energy, energy of zero-point vibrations, thermodynamic potential, and equation of state of solid molecular hydrogen in the volume range 10–60 a.u./atom. The calculations are carried out in the group approximation and third-order terms are included. It is shown that, for given values of the specific volume, allowance for the three-particle interactions reduces the energy and pressure compared with the pair approximation. Estimates are given of the precision and limits of validity of earlier pair-approximation calculations [V. P. Trubitsyn, Sov. Phys. Solid State **8**, 688 (1976); V. Magnasco and G. F. Musso, J. Chem. Phys. **47**, 1723 (1967); G. A. Neece, F. J. Rogers, and W. G. Hoover, J. Comput. Phys. **7**, 621 (1971); Hover *et al.* Phys. Earth Planet. Inter. **6**, 60 (1972); England *et al.* Phys. Rev. Lett. **32**, 758 (1974)].

PACS numbers: 31.70.Ks, 65.50.+m, 64.30.+t

In calculating the thermodynamic functions of the molecular phase of solid hydrogen, the main task is to determine the static energy of a crystal. This is usually done in the approximation of a pair interaction between the molecules forming the crystal.[1-5] When only the interaction between the nearest neighbors is taken into account in this approximation, the lattice energy per atom, measured from the energy of a free molecule, is given by

$$E=\frac{1}{2n}\sum_{\alpha=1}^{n}\left(\frac{1}{2}\sum_{j\neq\alpha}U_{\alpha,j}^{(2)}\right). \tag{1}$$

Here, n is the number of molecules in a unit cell; the index α labels the molecules in this cell and the index j identifies all the nearest neighbors of this molecule; $U_{\alpha,j}^{(2)}$ is the potential energy of the pair interaction between the molecules.

The expression (1) is the first (binary) term of the expansion of the energy as a series in groups of two, three, etc., molecules. It is shown by Nosanow[6] that the general form of this expansion for the bound-state energy of a system of N particles is

$$E=\sum_{\{k\}\subset N}\mathscr{E}_k(\{k\}). \tag{2}$$

The summation in Eq. (2) is carried out over all the k-particle subgroups of the original set of N particles.

The quantities $\mathscr{E}_k(\{k\})$ are found from the recurrence relationship

$$\mathscr{E}_k(\{k\})=\frac{(\psi_k(\{k\}),\ H_k(\{k\})\psi_k(\{k\}))}{(\psi_k(\{k\}),\ \psi_k(\{k\}))}-\sum_{m=1}^{k-1}\sum_{\{m\}\subset\{k\}}\mathscr{E}_m(\{m\}). \tag{3}$$

Here, $H_k(\{k\})$ is the Hamiltonian of a k-particle cluster:

$$H_k(\{k\})=-\frac{\hbar^2}{2M}\sum_{i=1}^{k}\nabla_i^2+U_k(\{k\}). \tag{4}$$

The mass of a single particle is denoted by M and the potential energy of the particles is $U_k(\{k\})$; $\psi_k(\{k\})$ is the wave function of the ground state of a k-particle cluster.

The potential energy of k molecules does not reduce to the sum of the energies of their pair interactions. In the approximation of adiabatic motion of the nuclei, we shall define it as the sum E_k of the electron energy and the energy of interaction between the nuclei at fixed positions. We are interested only in the static energy of the crystal so that we shall go to the limit $M\rightarrow\infty$. Then, $H_k=E_k$ is the potential energy of k molecules at equilibrium positions in the crystal lattice and Eq. (3) becomes

$$\mathscr{E}_k(\{k\})=E_k-\sum_{m=1}^{k-1}\sum_{\{m\}\subset\{k\}}\mathscr{E}_m(\{m\}).$$

Using Eq. (2) and allowing for the translational symmetry of crystals, we obtain the following expression for the energy of a crystal, calculated per atom:

$$E=\frac{1}{2n}\sum_{\alpha=1}^{n}\Big(\frac{1}{k}\sum_{\{k\}\subset N}\mathscr{E}_k(\{k\})\Big). \tag{5}$$

The summation in Eq. (5) is carried out over all the molecules forming a unit cell and over all the clusters containing such a molecule as a center.

We shall use the group expansion (5) to allow for three-particle clusters. In this case, the energy of a crystal, measured from the energy of a free molecule, is

$$E=\frac{1}{2n}\sum_{\alpha=1}^{n}\Big[\frac{1}{2}\sum_{j\neq\alpha}U_{\alpha,j}^{(2)}+\frac{1}{3}\sum_{\substack{j\neq\alpha\\ k\neq\alpha\\ k\neq j}}U_{\alpha,jk}^{(3)}\Big].$$

Here, $U_{\alpha,jk}^{(3)}$ is the three-particle contribution to the potential energy of the interaction between molecules α, j, and k, equal to the difference between the total energy of such a three-particle complex and the sum of one-particle energies of the molecules and energies of their pair interactions:

$$U_{\alpha,jk}^{(3)}=E_{\alpha,jk}-U_{\alpha,j}^{(2)}-U_{\alpha,k}^{(2)}-U_{j,k}^{(2)}-3E_{H_2}.$$

Similarly, the potential energy of the pair interaction is defined as the difference between the total energy of a pair of molecules and the sum of their one-particle energies:

$$U_{\alpha,j}^{(2)}=E_{\alpha,j}-2E_{H_2}.$$

The total energies of three- and two-particle clusters were found by applying the variational method to valence bonds with atomic $1S$ orbitals. The electron wave function of a system of three hydrogen molecules was represented by an expansion in terms of 37 basis functions, which were conveniently selected by the Rumer diagram method (see Appendix I). The whole set of basis functions included five covalent, twelve singly and twelve doubly polarized, and eight triply polarized structures. (Examples of diagrams for structures of each type are given in Appendix I.) The complete set of diagrams was then constructed by successive transpositions of the nuclei. An analytic expression for the wave function was a linear combination of the Slater determinants of the type

$$a\bar{b}c\bar{d}e\bar{f}$$
$$=\det\{a(1)\alpha(1)b(2)\beta(2)c(3)\alpha(3)d(4)\beta(4)e(5)\alpha(5)f(6)\beta(6)\},$$

where $a(1)=(\xi^3/\pi)^{1/2}\exp(-\xi|\mathbf{r}_1-\mathbf{r}_a|)$ is the Slater $1S$ function;

$$\alpha=\begin{pmatrix}1\\0\end{pmatrix},\quad \beta=\begin{pmatrix}0\\1\end{pmatrix}$$

are the eigenfunctions of the one-particle operator S_z with the eigenvalues of $+1/2$ and $-1/2$, respectively. For example, the covalent structure represented by the diagram (I.1) in Appendix I is associated with the wave function

$$\varphi_1=a\bar{b}c\bar{d}e\bar{f}-a\bar{b}c\bar{d}\bar{e}f-a\bar{b}\bar{c}de\bar{f}+a\bar{b}\bar{c}d\bar{e}f-$$
$$-\bar{a}bc\bar{d}e\bar{f}+\bar{a}bc\bar{d}\bar{e}f+\bar{a}b\bar{c}de\bar{f}-\bar{a}b\bar{c}d\bar{e}f.$$

Examples of analytic representation of the wave functions are also given in Appendix I.

In the state described by each of the functions φ_i, the z component and the total spin of the system of six electrons vanish, and

$$S_z\varphi_i=0,\quad S^2\varphi_i=0\quad (i=1,\ 2,\ldots,\ 37).$$

The wave function of a three-molecule complex is a linear combination of the φ_i functions. The coefficients in this combination are the variational parameters. The total energy of the complex was determined as the smallest root of the secular equation, obtained as a result of energy variation.

The pair interaction energy $U_{\alpha,j}^{(2)}$ was calculated similarly. The wave function of the complex of two molecules was described by a linear combination of ten basis functions. Their structure and analytic forms, expressed in terms of the Slater determinants, were as given in Appendix II. The energy calculations reduced to the solution of the variational problem.

The energy of a crystal was calculated for two types of lattice: the hcp lattice with the molecular axes parallel to the principal axis, and a lattice in which the centers of the molecules formed a face-centered cube and the axes were directed parallel to the principal diagonals of this cube (α-nitrogen structure). Each molecule in these lattices had 12 nearest neighbors. In the calculation of the three-particle interactions, account was taken only of those clusters in which not more than one distance between the centers of the molecules exceeded the distance between the nearest neighbors. These three-particle clusters, whose number was 150 in each lattice, dominated the correction due to the three-particle interaction. One could also show that, among these, the greatest contribution was made by 24 three-particle clusters, in which all three distances were equal to the distance between the nearest neighbors; this happened in spite of the fact that the number of such structures was only a small proportion of the total number of three-particle complexes considered. The symmetry conditions reduced the number of the various three-particle complexes to 8 in the hcp structure and to 12 in the α-nitrogen structure.

The parameter ξ—the effective nuclear charge—was assumed to be 1.193 atomic units (a.u.), and the distance between the nuclei in a molecule to be 1.4166 a.u., i.e., equal to the value corresponding to the minimum energy of a free molecule calculated in the approximation of the same bonds (one covalent and one polarized). In the case of the hcp lattice, the ratio c/a of the height of the cell c to the length of its base $\underline{a}$ was varied in the pair interaction approximation. The results of variation of the cell shape in the hcp lattice yielded:

v,	10	15	20	30	40	50	60
c/a:	2.00	1.92	1.88	1.84	1.80	1.78	1.78

Clearly, a reduction in the specific volume, i.e., an increase in pressure, causes the lattice to become elongated along the principal axis.

The static energies of a molecular hydrogen crystal, calculated by the above method in the pair and three-particle approximations, are plotted as a function of the specific volume in Figs. 1a (hcp lattice) and 1b (α-nitrogen lattice). The values of the energy per atom, measured from the energy of a free molecule, are given. For both lattices, the correction due to the three-particle interactions is negative and its absolute value rises with increase in the density of the crystal.

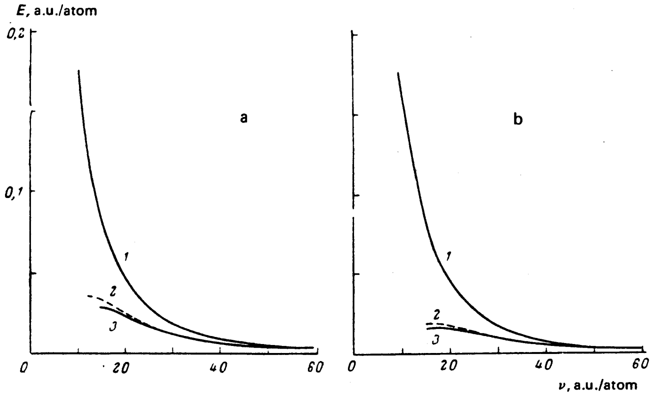


FIG. 1. Static energy of a crystal. (a) Hexagonal close-packed lattice: (1) pair interaction, nearest neighbors; (2) three-particle interaction allowing for second-nearest neighbors; (3) three-particle interaction allowing only for the nearest neighbors. (b) Lattice of the α-nitrogen type: (1) pair interaction, nearest neighbors; (2) three-particle interaction allowing for second-nearest neighbors; (3) three-particle interaction allowing only for the nearest neighbors.

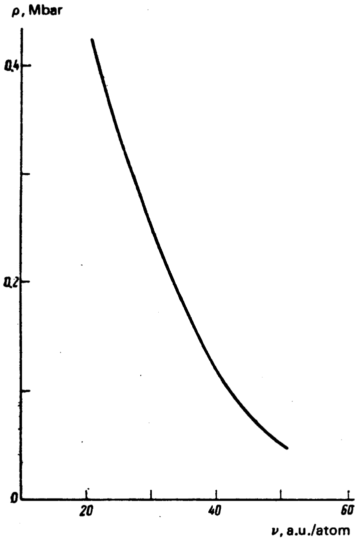


FIG. 2. Static component of the pressure for the hcp lattice.

In the range of specific volumes smaller than ~15 a.u./atom, the three-particle contribution rises so much that allowance for this contribution alters the sign of the isothermal compressibility. The inclusion of the zero-point vibrations does not basically alter the situation. Formally, the vanishing of the compressibility corresponds to the limit of stability of the investigated phase. However, it is clear that, in reality, the contribution of higher-order terms in the expansion (2) becomes considerable in the volume range under investigation and the adopted approach becomes invalid. In addition to a reduction in energy, this allowance for the three-particle interactions reduces the pressure. The static contribution to the pressure, calculated in the free-particle approximation, is shown in Fig. 2. The result applies to the hcp lattice; the corresponding dependence for the α-nitrogen lattice is exactly the same.

The calculated static energy of a crystal must be supplemented by the energy of zero-point vibrations. We calculated the latter in the Debye approximation, as was done by Abrikosov.[7] The total energy of a crystal, including that of zero-point vibrations, is plotted in Fig. 3 as a function of the specific volume of the hcp lattice. In the specific-volume range under consideration, the contribution of zero-point vibrations is relatively small and this justifies approximate allowance for these vibrations in the calculation of the thermodynamic functions.

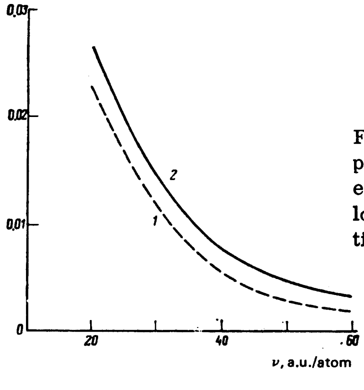


FIG. 3. Allowance for zero-point vibrations: (1) static energy; (2) energy with allowance for zero-point vibrations.

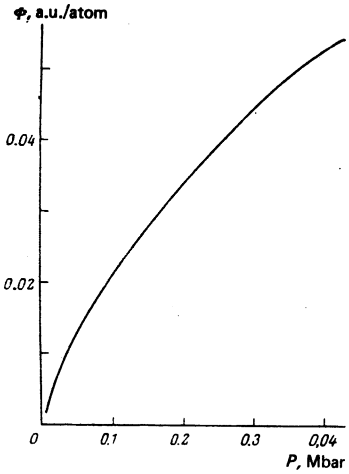
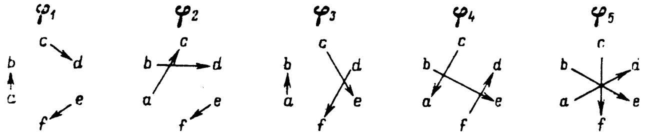


FIG. 4. Thermodynamic potential of molecular hydrogen (hcp lattice).

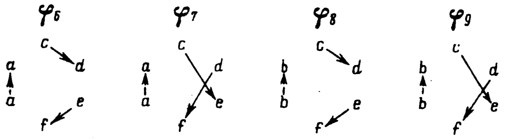
Figure 4 gives the thermodynamic potential of molecular hydrogen as a function of pressure at absolute zero. The result applies to the hcp lattice in the range of pressures in which the contribution of the ignored higher-order terms in Eq. (2) is of little significance.

APPENDIX I

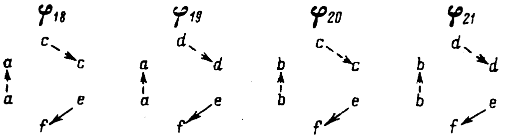
In a variational calculation of the energy of a system of three hydrogen molecules, the wave function was represented by a linear combination of 37 basis functions, which could conveniently be represented in the following form by means of the Rumer diagrams. We shall use the letters *a, b, c, d, e, f* to denote the protons in the three hydrogen molecules: *ab, cd,* and *ef.* A continuous arrow joining different letters (for example, *b* and *c*) means that the spin of electrons with orbitals centered on *b* and *c* have opposite directions. A dashed arrow joining identical letters (for example, $\underline{a}$ and $\underline{a}$) denotes that two electrons with opposite spins are located at the same nucleus $\underline{a}$. Examples of the basis functions of the three-molecular system are given below:



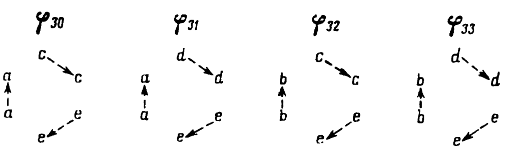
(I.1)



(I.2)



(I.3)



(I.4)

The covalent structures are represented by the diagrams (I.1), singly polarized by (I.2), doubly polarized by (I.3), and triply polarized by (I.4). Analytic forms of the wave functions, expressed in terms of the Slater determinants, are described by the formulas given below.

a) Covalent structures:

$$\varphi_1=a\bar{b}c\bar{d}e\bar{f}-a\bar{b}c\bar{d}\bar{e}f-a\bar{b}\bar{c}de\bar{f}+a\bar{b}\bar{c}d\bar{e}f$$
$$-\bar{a}bc\bar{d}e\bar{f}+\bar{a}bc\bar{d}\bar{e}f+\bar{a}b\bar{c}de\bar{f}-\bar{a}b\bar{c}d\bar{e}f,$$
$$\varphi_2=ab\bar{c}\bar{d}e\bar{f}-ab\bar{c}\bar{d}\bar{e}f-a\bar{b}\bar{c}de\bar{f}+a\bar{b}\bar{c}d\bar{e}f$$
$$\bar{a}bc\bar{d}e\bar{f}+\bar{a}bc\bar{d}\bar{e}f+\bar{a}\bar{b}cde\bar{f}-\bar{a}\bar{b}cd\bar{e}f,$$
$$\varphi_3=a\bar{b}cd\bar{e}\bar{f}-a\bar{b}c\bar{d}\bar{e}f-a\bar{b}\bar{c}de\bar{f}+a\bar{b}\bar{c}\bar{d}ef$$
$$-\bar{a}bcd\bar{e}\bar{f}+\bar{a}bc\bar{d}\bar{e}f+\bar{a}b\bar{c}de\bar{f}-\bar{a}b\bar{c}\bar{d}ef,$$
$$\varphi_4=ab\bar{c}d\bar{e}\bar{f}+a\bar{b}\bar{c}\bar{d}ef-a\bar{b}\bar{c}de\bar{f}-ab\bar{c}\bar{d}\bar{e}f$$
$$-\bar{a}bcd\bar{e}\bar{f}+\bar{a}bc\bar{d}\bar{e}f+\bar{a}\bar{b}cde\bar{f}-\bar{a}\bar{b}c\bar{d}ef,$$
$$\varphi_5=abc\bar{d}\bar{e}\bar{f}-ab\bar{c}\bar{d}\bar{e}f-a\bar{b}c\bar{d}e\bar{f}+a\bar{b}\bar{c}\bar{d}ef$$
$$-\bar{a}bcd\bar{e}\bar{f}+\bar{a}b\bar{c}d\bar{e}f+\bar{a}\bar{b}cde\bar{f}-\bar{a}\bar{b}\bar{c}def.$$

b) Singly polarized structures:

$$\varphi_6=a\bar{a}c\bar{d}e\bar{f}-a\bar{a}c\bar{d}\bar{e}f-a\bar{a}\bar{c}de\bar{f}+a\bar{a}\bar{c}d\bar{e}f,$$
$$\varphi_7=a\bar{a}cd\bar{e}\bar{f}-a\bar{a}c\bar{d}\bar{e}f-a\bar{a}\bar{c}de\bar{f}+a\bar{a}\bar{c}\bar{d}ef,$$
$$\varphi_8=b\bar{b}c\bar{d}e\bar{f}-b\bar{b}c\bar{d}\bar{e}f-b\bar{b}\bar{c}de\bar{f}+b\bar{b}\bar{c}d\bar{e}f,$$
$$\varphi_9=b\bar{b}cd\bar{e}\bar{f}-b\bar{b}c\bar{d}\bar{e}f-b\bar{b}\bar{c}de\bar{f}+b\bar{b}\bar{c}\bar{d}ef.$$

c) Doubly polarized structures:

$$\varphi_{18}=a\bar{a}c\bar{c}e\bar{f}-a\bar{a}c\bar{c}\bar{e}f,\quad \varphi_{19}=a\bar{a}d\bar{d}e\bar{f}-a\bar{a}d\bar{d}\bar{e}f,$$
$$\varphi_{20}=b\bar{b}c\bar{c}e\bar{f}-b\bar{b}c\bar{c}\bar{e}f,\quad \varphi_{21}=b\bar{b}d\bar{d}e\bar{f}-b\bar{b}d\bar{d}\bar{e}f.$$

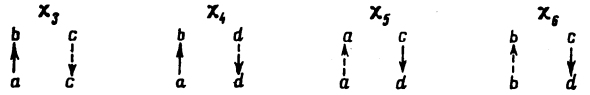
d) Triply polarized structures:

$$\varphi_{30}=a\bar{a}c\bar{c}e\bar{e},\quad \varphi_{31}=a\bar{a}d\bar{d}e\bar{e},$$
$$\varphi_{32}=b\bar{b}c\bar{c}e\bar{e},\quad \varphi_{33}=b\bar{b}d\bar{d}e\bar{e}.$$

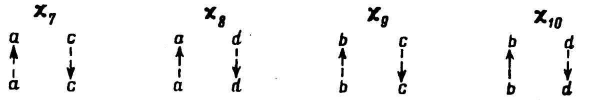
APPENDIX II

The wave functions of two-molecule complexes were represented by linear combinations of 10 basis functions, corresponding to the following diagrams:

(II.1)



(II.2)



(II.3)

Covalent structures correspond to the diagrams (II.1), singly polarized structures to (II.2), and doubly polarized to (II.3).

We shall also give the analytic formulas corresponding to these diagrams.

a) Covalent structures:

$$\chi_1=a\bar{b}c\bar{d}-a\bar{b}\bar{c}d-\bar{a}bc\bar{d}+\bar{a}b\bar{c}d,$$
$$\chi_2=ab\bar{c}\bar{d}-a\bar{b}\bar{c}d-\bar{a}bc\bar{d}+\bar{a}\bar{b}cd.$$

b) Singly polarized structures:

$$\chi_3=a\bar{b}c\bar{c}-\bar{a}bc\bar{c},\quad \chi_4=a\bar{b}d\bar{d}-\bar{a}bd\bar{d},$$
$$\chi_5=a\bar{a}c\bar{d}-a\bar{a}\bar{c}d,\quad \chi_6=b\bar{b}c\bar{d}-b\bar{b}\bar{c}d.$$

c) Doubly polarized structures.

$$\chi_7=a\bar{a}c\bar{c},\quad \chi_8=a\bar{a}d\bar{d},$$
$$\chi_9=b\bar{b}c\bar{c},\quad \chi_{10}=b\bar{b}d\bar{d}.$$

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Translated by A. Tybulewicz

Magnetic properties of disordered media

A. F. Andreev

Institute of Physical Problems, USSR Academy of Sciences

(Submitted 21 September 1977)
Zh. Eksp. Teor. Fiz. 74, 786–797 (February 1978)

It is shown that in spatially disordered magnetic systems three macroscopically different types of magnetically disordered exchange structures are possible. Besides a disordered ferromagnet and a completely random structure of the spin-glass type, a structure corresponding to a disordered antiferromagnet with three mutually perpendicular antiferromagnetic moments is possible. Nonlinear equations are found that describe the dynamical properties of spin glasses and disordered ferromagnets and antiferromagnets with allowance for the external magnetic field and relativistic interactions. The spin-wave spectrum and magnetic-resonance frequencies are calculated.

PACS numbers: 75.30.Ds, 75.30.Et

This paper is devoted to a macroscopic analysis of the magnetic properties of spatially disordered media, i.e., systems in which the spatial distribution of the magnetic atoms is, on the average, homogeneous and isotropic. We are concerned here with amorphous substances containing magnetic atoms or with weak solutions of magnetic atoms in nonmagnetic crystals. We shall assume that the appearance of one magnetic structure or another in the substances under consideration is due principally to exchange forces considerably greater than the relativistic interactions.

A spatially disordered system can be completely ordered magnetically. The only such case is complete ferromagnetic ordering of the spins of the magnetic atoms. Any other magnetic order is obviously incompatible with spatial disorder. The macroscopic properties of such a ferromagnet do not differ from the properties of ordinary crystalline ferromagnets and are described by the Landau-Lifshitz equation.[1]

In recent times, disordered systems with magnetic structures of a different type have been widely investigated (see the reviews[2,3]). These, firstly, are the so-called spin glasses (see Ref. 2), in which not only the positions but also the directions of the spins of the different atoms are randomly distributed. In addition, there exist systems (see Ref. 3) possessing a finite spontaneous magnetization whose value at zero temperature, however, differs substantially from the nominal value. The state of such a disordered ferromagnet is analogous to the state of a spin glass in an external magnetic field. There is partial ferromagnetic order superimposed on a fairly random (in general) distribution of spin orientations.

Below, on the basis of the symmetry arguments developed in the work of Marchenko and the author,[4] all the theoretically possible macroscopically different types of such partial order will be found. It turns out that, apart from the disordered ferromagnet, there exists only one other possible structure—a disordered antiferromagnet characterized by three mutually perpendicular antiferromagnetic moments.

The dynamical properties of disordered magnetic media can be described macroscopically in a manner analogous to the way in which ordinary amorphous solids are described by the theory of elasticity. In this case the analog of the spatial-displacement vector that appears in elasticity theory is a rotation of all the spins through the same angle. The exchange energy does not change under such a rotation. The change of energy is determined, therefore, by the time and space derivatives of the rotation angles, which are analogous to the velocity of the medium and the deformation. The description of the spin dynamics by means of rotation angles has been used in the study of the magnetic properties of the superfluid phases of He^3 [5,6] and of crystalline[7] and disordered[8] magnets. There is, however, an essential difference between elasticity theory and magnetic dynamics. Unlike spatial displacements, different rotations do not, generally speaking, commute. Therefore, the equations of magnetic dynamics are nonlinear even for small velocities and deformations. Below we shall derive the dynamical equations for all three types of disordered magnetic structures, i.e., for spin glasses and disordered ferromagnets and antiferromagnets. For our purposes it will be convenient to use the Lagrangian formalism applied by Maki[6] to investigate the magnetic properties of liquid He^3-*B* and by Dzyaloshinskiĭ and Kukharenko[9] for crystalline magnets. The spin-glass case is the simplest. The