Molecular hydrogen crystal in the multiparticle approximation

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(Submitted 2 August 1982)
Zh. Eksp. Teor. Fiz. **84**, 778–782 (February 1983)

The valence-band method is used to calculate the energy of an n-atom ($n = 38$) system of hydrogen molecules located in an α-nitrogen-type lattice of solid molecular hydrogen at densities in the region of the metal-insulator phase transition. The limits of applicability of the approximation based on the cluster expansion of the energy, with allowance for three-particle interactions, are estimated.

PACS numbers: 31.70.Ks

An important factor in the problem of hydrogen metallization under strong compression is the properties of the molecular phase that is the precursor of the metallic state at low pressures. It is precisely the large uncertainty in the equation of state of the molecular crystal which causes for the most part large discrepancies in the predicted pressure of the transition from the molecular to the metallic phase. The general assumption in the study of the molecular phase is the paired character of the interaction of the molecules that make up the crystal. This leads to the four-electron problem, the solution of which is the subject of many papers.[1–7] Since, however the proposed phase transition takes place in the region of an appreciable compression of the crystal (its density increases tenfold compared with the crystal density at zero pressure), it becomes clear that the use of the approximation of pair interaction of "bare" molecules in the entire interval of the investigated densities is not sufficiently well founded. At densities of interest for the investigation of the phase transition, a larger role is assumed by the multiparticle effects. It was indicated in Ref. 6 that the non-additive three-particle interaction of hydrogen molecules increases when the distances between them are decreased. In the study of the non-additive interaction of three hydrogen molecules one encounters a six-electron problem. The energy of a molecular hydrogen crystal having the structure of α nitrogen was calculated at zero temperature in Ref. 8, using the cluster approximation and considering clusters consisting of three molecules. The energy of a three-molecule cluster was calculated by the method of valence bonds with account taken of all the covalent and of the most important singly, doubly, and triply ionized structures. It was shown that allowance for the non-additive three-particle interaction leads to a lower energy than the pair-interaction approximation, and this leads correspondingly to a "softer" equation of state, i.e., to weaker increase of pressure with increasing density than in the case of pair interaction. At specific volumes less than 20 a.u./atom, an important role is assumed in the cluster expansion by clusters consisting of a large number of molecules, and this limits the applicability of the three-particle-interaction approximation. The present paper deals with the molecular phase of hydrogen at high densities, including the indicated region where multiparticle effects become substantial.

MULTIELECTRON WAVE FUNCTION

The considered region of the specific volume (10–50 a.u./atom) corresponds in the case of a lattice of the α-nitrogen type to nearest neighbors separated by distances larger by 2–4 molecule sizes. We construct therefore the multiparticle wave function with localized atomic orbitals, using the valence-bond method. In this case the relatively large intermolecular distances, compared with the distance between the nuclei in the molecule, allows us to confine ourselves to only one valence structure corresponding to the bonds produced between electrons localized on one molecule, and neglect the bonds between electrons of different molecules. The normalized electron wave function of the ground state can then be written in the form

$$\Psi(1,2,\ldots,2N= \sum_{s_1=\pm1,\ldots,s_N=\pm1} (-1)^{n_+}$$

$$\times \det\{A_1^{s_1}B_1^{-s_1}A_2^{s_2}B_2^{s_2}\ldots A_N^{s_N}B_N^{-s_N}\};$$

$$A_1^{+1}=\Phi(|\mathbf{r}-\mathbf{A}_1|)\begin{pmatrix}1\\0\end{pmatrix},\quad B_1^{-1}=\varphi(|\mathbf{r}-\mathbf{B}_1|)\begin{pmatrix}0\\1\end{pmatrix}. \quad (1)$$

Here A_1^{+1} is the spin orbital localized on the nucleus A of molecule 1 and corresponding to a spin z-projection equal to $+\frac{1}{2}$; B_1^{-1} is a spin orbital localized on nucleus B of molecule 1, with a spin z-projection equal to $-\frac{1}{2}$; n_+ is the number of s equal to $+1$ (the number of spins on the orbitals centered on the nuclei A and directed upward); N is the number of molecules.

To simplify the calculations that follow, we consider a system of orbitals $\{\Phi_i, \varphi_i\}$ ($i = 1,2,\ldots, N$) such that

$$\langle\Phi_i|\Phi_j\rangle=\langle\Phi_i|\varphi_j\rangle=\langle\varphi_i|\varphi_j\rangle=0,\quad i\neq j,$$
$$\langle\Phi_i|\varphi_i\rangle\neq0. \quad (2)$$

This means that the atomic orbitals belonging to different valence bonds are orthogonal to one another, but the overlap integral of the orbitals of one bond differs from zero. This corresponds to the known Harley-Lennard-Jones-Pople approximation for molecules.

0038-5646/83/020449-04$04.00

CONSTRUCTION OF SYSTEM OF ATOMIC ORBITALS

We shall construct the required system of atomic orbitals $\{\Phi_i, \varphi_i\}$ out of initial Slater 2s orbitals, which we divide into two groups, $\{a_i\}$ and $\{b_i\}$, corresponding in the α-nitrogen lattice to the lower and upper nuclei in the molecule. We orthogonalize any one of these groups, say $\{a_i\}$, by Löwdin's method. We obtain a system of orbitals $\{\Phi_i\}$ localized on the lower nuclei of the molecules:

$$\Phi_i = \sum_{j=1}^{N} D_{ij} a_j. \tag{3}$$

Here D is the square root of the matrix inverse to the overlap matrix a of the atomic orbitals $\{a_i\}$:

$$(\Delta_a)_{ij} = \langle a_i | a_j \rangle, \quad D = \Delta_a^{-1/2}.$$

At the specific-volume values considered by us, the off-diagonal elements of the matrix Δ_a are small compared with the diagonal ones. The system of orbitals $\{\Phi_i\}$ is orthonormalized:

$$\langle \Phi_i | \Phi_j \rangle = \delta_{ij}. \tag{4}$$

We form now a second system of orbitals $\{\varphi_i\}$ localized on the upper nuclei of the molecules. This system of orbitals should satisfy the conditions

$$\langle \varphi_i | \varphi_j \rangle = \delta_{ij}, \quad \langle \varphi_i | \Phi_j \rangle = 0, \quad i \neq j, \quad \langle \varphi_i | \Phi_i \rangle \neq 0. \tag{5}$$

We shall construct the system $\{\varphi_i\}$ out of $\{b_i\}$ and $\{\Phi_i\}$. To this end we seek φ_1 in the form

$$\varphi_1 = b_1 + \sum_{i \neq 1} A_{1,i} \Phi_i. \tag{6}$$

We have left out of the sum the term with $i = 1$, i.e., the function a_1 does not take part in the formation of the orbital φ_1. The orthogonality condition (5) for the function φ_1 takes the following form:

$$\langle \varphi_1 | \Phi_j \rangle = 0, \quad j \neq 1. \tag{7}$$

Substituting here φ_1 from (6) we obtain for $j \neq 1$

$$\langle b_1 | \Phi_j \rangle + \sum_{i \neq 1} A_{1,i} \langle \Phi_i | \Phi_j \rangle = 0.$$

Hence, since the functions $\{\Phi_i\}$ are orthonormal, we obtain

$$A_{1,j} = -\langle b_1 | \Phi_j \rangle, \quad j \neq 1. \tag{8}$$

We construct now the functions $\varphi_1, \varphi_2, \ldots, \varphi_k$. We seek the function φ_{k+1} in the form

$$\varphi_{k+1} = b_{k+1} + \sum_{i=1}^{k} B_{k+1,i} \varphi_i + \sum_{m \neq k+1} A_{k+1,m} \Phi_m. \tag{9}$$

The following system of orthogonality relation holds for the function φ_{k+1}

$$\langle \varphi_{k+1} | \varphi_j \rangle = 0, \quad j = 1, 2, \ldots, k, \tag{10}$$

$$\langle \varphi_{k+1} | \Phi_j \rangle = 0, \quad j = 1, 2, \ldots, N, \quad j \neq k. \tag{11}$$

The condition (10), with allowance for the function φ_{k+1} in the form (9) yields ($j = 1, 2, \ldots, k$)

$$\langle b_{k+1} | \varphi_j \rangle + \sum_{i=1}^{k} B_{k+1,i} \langle \varphi_i | \varphi_j \rangle + \sum_{m \neq k+1} A_{k+1,m} \langle \Phi_m | \varphi_j \rangle = 0.$$

Hence, recognizing that for $j = 1, 2, \ldots, k$

$$\langle \varphi_i | \varphi_j \rangle = \langle \varphi_j | \varphi_j \rangle \delta_{ij}, \quad \langle \Phi_m | \varphi_j \rangle = \langle \Phi_j | \varphi_j \rangle \delta_{mj},$$

we obtain

$$\langle b_{k+1} | \varphi_j \rangle + B_{k+1,j} \langle \varphi_j | \varphi_j \rangle + A_{k+1,j} \langle \Phi_j | \varphi_j \rangle = 0, \quad (j = 1, 2, \ldots, k). \tag{12}$$

Relation (11) yields for $j \neq k + 1$

$$\langle b_{k+1} | \Phi_j \rangle + \sum_{i=1}^{k} B_{k+1,i} \langle \varphi_i | \Phi_j \rangle + \sum_{m \neq k+1} A_{k+1,m} \langle \Phi_m | \Phi_j \rangle = 0. \tag{13}$$

For $j = 1, 2, \ldots, k$ these equations take the form

$$\langle b_{k+1} | \Phi_j \rangle + B_{k+1,j} \langle \varphi_j | \Phi_j \rangle + A_{k+1,j} = 0. \tag{14}$$

Solving (12) and (14) simultaneously, we obtain ($j = 1, 2, \ldots, k$)

$$A_{k+1,j} = \frac{\langle b_{k+1} | \varphi_j \rangle \langle \varphi_j | \Phi_j \rangle - \langle b_{k+1} | \Phi_j \rangle \langle \varphi_j | \varphi_j \rangle}{\langle \varphi_j | \varphi_j \rangle - \langle \varphi_j | \Phi_j \rangle \langle \Phi_j | \varphi_j \rangle},$$
$$B_{k+1,j} = \frac{\langle b_{k+1} | \Phi_j \rangle \langle \Phi_j | \varphi_j \rangle - \langle b_{k+1} | \varphi_j \rangle}{\langle \varphi_j | \varphi_j \rangle - \langle \varphi_j | \Phi_j \rangle \langle \Phi_j | \varphi_j \rangle}. \tag{15}$$

Equations (13) reduce for $j = k + 2, k + 3, \ldots, N$ to

$$\langle b_{k+1} | \Phi_j \rangle + A_{k+1,j} = 0.$$

From this we get

$$A_{k+1,j} = -\langle b_{k+1} | \Phi_j \rangle, \quad j = k+2, k+3, \ldots, N. \tag{16}$$

Equation (9) together with (15) and (16) yields a recurrence relation for the function φ_{k+1} in terms of the functions $\varphi_1, \varphi_2, \ldots, \varphi_k$ and the function $\{\Phi_i\}$. We now express the functions $\{\varphi_i\}$ in terms of the initial systems of functions $\{a_i\}$ and $\{b_i\}$:

$$\varphi_i = \sum_{j=1}^{N} P_{ij} a_j + \sum_{l=1}^{N} Q_{il} b_l. \tag{17}$$

Here P and Q are still undetermined $N \times N$ matrices. We introduce recurrence relations for the elements of the matrices P and Q. From (6) we have

$$\varphi_1 = b_1 + \sum_{i \neq 1} A_{1,i} \Phi_i = b_1 + \sum_{j=1}^{N} \sum_{i \neq 1} A_{1,i} D_{ij} a_j.$$

It follows therefore that

$$P_{1j} = \sum_{i \neq 1} A_{1,i} D_{ij}, \quad j = 1, 2, \ldots, N,$$
$$Q_{11} = 1, \quad Q_{1j} = 0, \quad j = 2, \ldots, N. \tag{18}$$

Substituting φ_i from (17) in expression (9) for φ_{k+1}, we obtain

$$\varphi_{k+1}=\sum_{j=1}^{N}\Bigl(\sum_{i=1}^{k}B_{k+1,i}P_{ij}+\sum_{\substack{m=1\\ m\neq k+1}}^{N}A_{k+1,m}D_{mj}\Bigr)a_j$$

$$+\sum_{\substack{l=1\\ l\neq k+1}}^{N}\Bigl(\sum_{i=1}^{k}B_{k+1,i}Q_{il}\Bigr)b_l+\Bigl(1+\sum_{i=1}^{k}B_{k+1,i}Q_{i,k+1}\Bigr)b_{k+1}.$$

We have thus obtained the following recurrence relations for the elements of the matrices P and Q:

$$P_{k+1,j}=\sum_{i=1}^{k}B_{k+1,i}P_{ij}+\sum_{\substack{m=1\\ m\neq k+1}}^{N}A_{k+1,m}D_{mj},$$

$$Q_{k+1,j}=\sum_{i=1}^{k}B_{k+1,i}Q_{ij},\quad j\neq k+1,\tag{19}$$

$$Q_{k+1,k+1}=1+\sum_{i=1}^{k}B_{k+1,i}Q_{i,k+1}.$$

Together with expressions (18) for the elements of the first rows of P_{ij} and Q_{ij}, they enable us to determine the elements of all the rows of the matrices P and Q from the coefficients A and B. In our case, in accordance with Eqs. (15), these coefficients are expressed in terms of the functions $\{\Phi_i\}$ and $(\varphi_i\}$. We express them in terms of the initial sets of orbitals $\{a_i$ and $\{b_i\}$. We introduce in addition to the overlap matrix Δ_a of the orbitals $\{a_i\}$ the overlap matrix Δ_{ab} of the orbitals $\{a_i\}$ and $\{b_i\}$ as well as the matrix Δ_b for the overlap of the orbitals $\{b_i\}$ with one another, so that

$$(\Delta_{ab})_{ij}=\langle a_i|b_j\rangle,\qquad(\Delta_b)_{ij}=\langle b_i|b_j\rangle,\tag{20}$$

Substituting in (15) the functions $\{\Phi_i\}$ and $\{\Phi_i\}$ from (3) and (17), respectively, we obtain for $j=1, 2,\ldots, k$

$$A_{k+1,j}=\frac{[P\Delta_{ab}+Q\Delta_b]_{j,k+1}[D(\Delta_aP^{\mathrm{T}}+\Delta_{ab}Q^{\mathrm{T}})]_{jj}-[D\Delta_{ab}]_{j,k+1}[P\Delta_aP^{\mathrm{T}}+2P\Delta_{ab}Q^{\mathrm{T}}+Q\Delta_bQ^{\mathrm{T}}]_{jj}}{[P\Delta_aP^{\mathrm{T}}+2P\Delta_{ab}Q^{\mathrm{T}}+Q\Delta_bQ^{\mathrm{T}}]_{jj}-\{[D(\Delta_aP^{\mathrm{T}}+\Delta_{ab}Q^{\mathrm{T}})]_{jj}\}^2},\tag{21}$$

$$B_{k+1,j}=\frac{[D\Delta_{ab}]_{j,k+1}[D(\Delta_aP^{\mathrm{T}}+\Delta_{ab}Q^{\mathrm{T}})]_{jj}-[P\Delta_{ab}+Q\Delta_b]_{j,k+1}}{[P\Delta_aP^{\mathrm{T}}+2P\Delta_{ab}Q^{\mathrm{T}}+Q\Delta_bQ^{\mathrm{T}}]_{jj}-\{[D(\Delta_aP^{\mathrm{T}}+\Delta_{ab}Q^{\mathrm{T}})]_{jj}\}^2},$$

and for $j=k+2,\ldots, N$

$$A_{k+1,j}=-[D\Delta_{ab}]_{j,k+1}.\tag{21'}$$

Here P^T is the transpose of the matrix P. It is necessary to add to Eqs. (21) the value of A_{1j}:

$$A_{1,j}=-[D\Delta_{ab}]_{j1},\quad j\neq1,\tag{22}$$

Now, knowing P, Q, A and B on the k-th step, we determine on the next $(k+1)$st step first A and B from (21), and from them next P and Q. It will be more convenient to use hereafter the functions $\{\varphi_i$ normalized, just as $\{\Phi_i\}$, to unity:

$$\langle\varphi_i|\varphi_j\rangle=\delta_{ij}.$$

We normalize the functions $\{\varphi_i\}$ by introducing in place of the matrices P and Q the matrices T^α $(=1,2)$:

$$T_{ij}^1=P_{ij}/([P\Delta_aP^{\mathrm{T}}+2P\Delta_{ab}Q^{\mathrm{T}}+Q\Delta_bQ^{\mathrm{T}}]_{ii})^{1/2},$$

$$T_{ij}^2=Q_{ij}/([P\Delta_aP^{\mathrm{T}}+2P\Delta_{ab}Q^{\mathrm{T}}+Q\Delta_bQ^{\mathrm{T}}]_{ii})^{1/2}.\tag{23}$$

We also introduce a common symbol c^α $(\alpha=1,2)$ for the orbitals $\{a_i\}$ and $\{b_i\}$:

$$a_i=c_i^1,\quad b_i=c_i^2.\tag{24}$$

The orbitals $\{\varphi_i\}$ can now be written in the form:

$$\varphi_i=T_{ij}^\alpha c_j^\alpha\tag{25}$$

(summation is carried out over α and j).

ENERGY OF FACE CENTERED CUBIC LATTICE

The Hamiltonian of a system of N hydrogen molecules has in the Born-Oppenheimer approximation with secured nuclei the usual form

$$H=-\frac{1}{2}\sum_{i=1}^{2N}\nabla_i^2-\sum_{i,j}|\mathbf{r}_i-\mathbf{R}_j|^{-1}+\frac{1}{2}\sum_{i,j}{}'|\mathbf{r}_i-\mathbf{r}_j|^{-1}$$

$$+\frac{1}{2}\sum_{i,j}{}'|\mathbf{R}_i-\mathbf{R}_j|^{-1},\tag{26}$$

where $\mathbf{r}_i$ and $\mathbf{R}_i$ are the coordinates of the electrons and of the nuclei ($i=1, 2,\ldots, 2N$). The nuclei make up a lattice of α-nitrogen type, in which the centers of the molecules constitute a face-centered cube, and the molecular axes are directed along the body diagonals of this cube.

Calculating in the usual manner the mean value of the Hamiltonian (26) in a state with wave function (1), $E=\langle\Psi|H|\Psi\rangle/\langle\Psi|\Psi\rangle$, we obtain

$$E=E_1+E_2^d+E_2^{ex},\tag{27}$$

where

$$E_1=\sum_{i=1}^{N}(1+S_i^2)^{-1}\{D_{im}[D_{in}(a_m|a_n)+2S_iT_{in}^\alpha(a_m|c_n^\alpha)]$$
$$+T_{im}^\alpha T_{in}^\beta(c_m^\alpha|c_n^\beta)$$
$$+D_{im}T_{iq}^\beta[D_{in}T_{ip}^\alpha(a_ma_n|c_p^\alpha c_q^\beta)$$
$$+T_{in}^\alpha D_{ip}(a_mc_n^\alpha|a_pc_q^\beta)]\}\tag{28}$$

is the sum of the renormalized energies of the individual molecules,

$$E_2^d=\sum_{i<j}(1+S_i^2)^{-1}(1+S_j^2)^{-1}\{D_{im}D_{in}[D_{jp}D_{jq}(a_ma_n|a_pa_q)$$
$$+T_{jp}^\gamma T_{jq}^\delta(a_ma_n|c_p^\gamma c_q^\delta)]+T_{im}^\alpha T_{in}^\beta[D_{jp}D_{jq}(c_m^\alpha c_n^\beta|a_pa_q)$$
$$+T_{jp}^\gamma T_{jq}^\delta(c_m^\alpha c_n^\beta|c_p^\gamma c_q^\delta)]+2S_iD_{im}T_{in}^\beta[D_{jp}D_{jq}(a_mc_n^\beta|a_pa_q)$$
$$+T_{jp}^\gamma T_{jq}^\delta(a_mc_n^\beta|c_p^\gamma c_q^\delta)]+2S_jD_{jm}T_{jn}^\beta[D_{ip}D_{iq}(a_mc_n^\beta|a_pa_q)$$
$$+T_{ip}^\gamma T_{iq}^\delta(a_mc_n^\beta|c_p^\gamma c_q^\delta)]+4S_iS_jD_{im}T_{in}^\beta D_{jp}T_{jq}^\delta(a_mc_n^\beta|a_pc_q^\delta)\}\tag{29}$$

is the renormalized energy of the direct interaction of the molecules, and

$$E_2^{ex}=-\frac{1}{2}\sum_{i<j}(1+S_i^2)^{-1}(1+S_j^2)^{-1}\{D_{im}D_{ip}[D_{jn}D_{jq}(a_ma_n|a_pa_q)$$
$$+T_{jn}^\beta T_{jq}^\delta(a_mc_n^\beta|a_pc_q^\delta)]+T_{im}^\alpha T_{ip}^\gamma[D_{jn}D_{jq}(c_m^\alpha a_n|c_p^\gamma a_q)$$
$$+T_{jn}^\beta T_{jq}^\delta(c_m^\alpha c_n^\beta|c_p^\gamma c_q^\delta)]+2S_iD_{im}T_{iq}^\delta[D_{jn}D_{jp}(a_ma_n|a_pc_q^\delta)$$
$$+T_{jn}^\beta T_{jp}^\gamma(a_mc_n^\beta|c_p^\gamma c_q^\delta)]+2S_jD_{jm}T_{jq}^\delta[D_{in}D_{ip}(a_ma_n|a_pc_q^\delta)$$
$$+T_{in}^\beta T_{ip}^\gamma(a_mc_n^\beta|c_p^\gamma c_q^\delta)]+2S_iS_jD_{im}T_{iq}^\delta[T_{jn}^\beta D_{jp}(a_mc_n^\beta|a_pc_q^\delta)$$
$$+D_{jn}T_{jp}^\gamma(a_ma_n|c_p^\gamma c_q^\delta)]\}\tag{30}$$

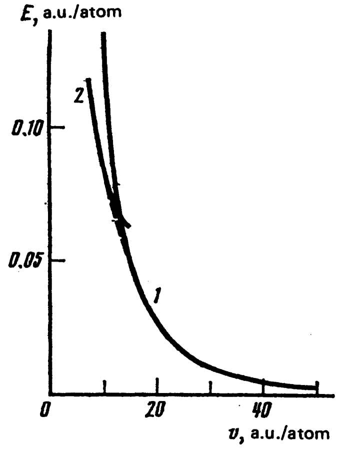

FIG. 1. Energy of molecular and atomic phases of solid hydrogen: 1—energy of n-atom cluster of molecular hydrogen ($n = 39$); 2—energy of atomic phase 11; dashes—common tangent to the two curves.

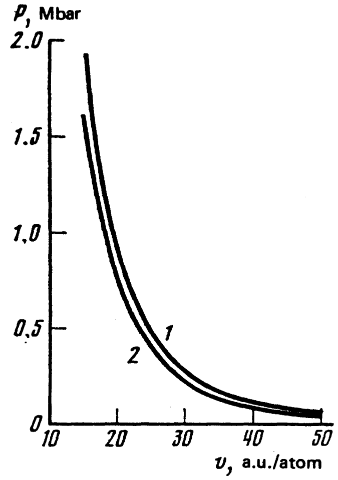

FIG. 2. Dependence of the pressure of the molecular phase in the volume: 1—result of present paper, 2—experimental curve.[12]

is the renormalized exchange interaction.

In (28)–(30) summation over repeated indices is implied throughout.

Here

$$S_i = \langle \Phi_i | \varphi_i \rangle = D_{im} T_{in}{}^{\beta} \langle a_m | c_n{}^{\beta} \rangle$$

is the overlap integral of the orbitals Φ_i and φ_i

$$(a_m | a_n) = \left\langle a_m \left| -\frac{1}{2} \nabla^2 - \sum_j |\mathbf{r} - \mathbf{R}_j|^{-1} \right| a_n \right\rangle$$

is the single-electron matrix element, and

$$(a_m a_n | a_p a_q) = \int a_m(1) a_n(1) r_{12}{}^{-1} a_p(2) a_q(2) d^3 1 \, d^3 2$$

is the two-electron matrix element.

In this paper the initial atomic orbitals $\{a_i\}$ and $\{b_i\}$ were taken in the form of a linear combination of two Gaussian 1s functions with the expansion coefficients obtained in Ref. 9. Equations (28)–(30) were used to calculate the energy of a system of molecules located in two coordination spheres ($N = 19$). The results of the calculation of the energy as a function of the specific volume of the crystal is shown in Fig. 1. The same figure shows the metallic-phase energy calculated by perturbation theory,[10,11] and this enables us to estimate the pressure of the transition from the molecular into the metallic phase, namely ~2 Mbar. The binding energies of both phases were reckoned from the free molecules and referred to one atom. The energy of the molecular phase, obtained in the multiparticle approximation considered here, is in good agreement with the energy calculated in Ref. 8 in the three-particle-interaction approximation at a specific volume of the crystal larger than 30 a.u./atom. We can therefore conclude that the description of molecular solid hydrogen is adequately described in this region by a cluster expansion with three-molecular clusters taken into account. Numerical differentiation of the dependence of the energy on the volume in Fig. 1 yielded the dependence of the pressure on the volume. The corresponding curve is shown in Fig. 2 together with a number of points obtained in a recent precision experiment[12] in which molecular hydrogen was strongly compressed at low temperature ($T = 5$ K). The two curves are in good agreement.

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Translated by J. G. Adashko