The Lattice Energy of Solid CO2

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It is possible to account for the measured sublimation heat of solid CO₂ by calculating the attraction energy according to London's theory of van der Waals forces; or, when the second approximation of the attractive forces is included, by considering also the electrostatic energy due to the dipole moments and the repulsive energy, the latter being deduced from the equilibrium condition. For the calculation of the electrostatic energy the picture of point charges with definite distances seems preferable to that of short dipoles.

IN the course of other investigations we became interested in the question as to what extent an application of the London theory of van der Waals forces and the present representation of repulsion forces would account for the lattice energies of molecular lattices with linear triatomic molecules such as CO₂ and N₂O. These substances crystallize in cubic face-centered lattices, a type which has been treated by London[1] for simple diatomic molecules. A very rough orienting calculation by treating the molecules as if concentrated in one point, gave the right order of magnitude for the sublimation heats which represent the lattice energies. The values which we obtained for CO₂ will be given in Table I in connection with the more exact calculations.

CALCULATION IN FIRST APPROXIMATION

The CO₂ gas crystallizes at very low temperatures in a cubic face-centered lattice, as mentioned above, with the following distances: $C-C$ distance between nearest neighbors $g=3.917A$, $C-O$ distance in one molecule $f=1.12A$ and $C-C$ distance between neighbored corners of the cube $a=5.54A$.[2] Those molecules CO₂ located with their C atoms in the corners of the cube point in the direction of the diagonal going through the centers of the cubes. They are all parallel to each other. Those molecules, however, which are located with their C atoms in face centers point to the centers of the edges of the cube. (See Fig. 1.)

London has derived a simple expression for the energy of a molecular lattice considering only dipole-dipole interactions:

$$\Phi = -\frac{CN_L}{2}\sum_i \frac{1}{R_i{}^6} \quad \text{with} \quad C=\tfrac{3}{4}\alpha^2 h\nu, \quad (1)$$

where N_L is Loschmidt's number, R_i are the distances from one fixed atom to all the other atoms in the lattice, α the polarizability of the atom and $h\nu$ a characteristic energy amount, which corresponds to the principal specific frequency ν taken from the dispersion equation of the particle in question (or about the ionization energy if the dispersion equation is not known). We extend the summation over the distances smaller than the edge of the unit cube calculating the rest by integration.

In the case of CO₂ it seemed plausible to assume that practically only the O atoms contribute to the polarizability, the α value for the C atom in the center being much smaller than for the O atoms.[3] It seems furthermore plausible that in this two-center model for the CO₂ molecule the polarizability of the centers should not be located in the nucleus of the O atoms but should be a little shifted toward the C atom. For this shift there are several assumptions possible which are all more or less arbitrary. In order to remove this arbitrariness we have applied Silberstein's[4] theory to our model although we are aware that this theory is not ideal. But besides removing the arbitrariness of locating the polarizabilities, Silberstein's theory has the other advantage of taking into account

[1] F. London, Zeits. f. physik. Chemie **B11**, 222 (1930).
[2] W. H. Keesom and I. W. L. Köhler, Physica **1**, 167 and 655 (1934).
[3] See for similar considerations F. V. Lenel, Zeits. f. physik. Chemie **B23**, 379 (1933), p. 390.
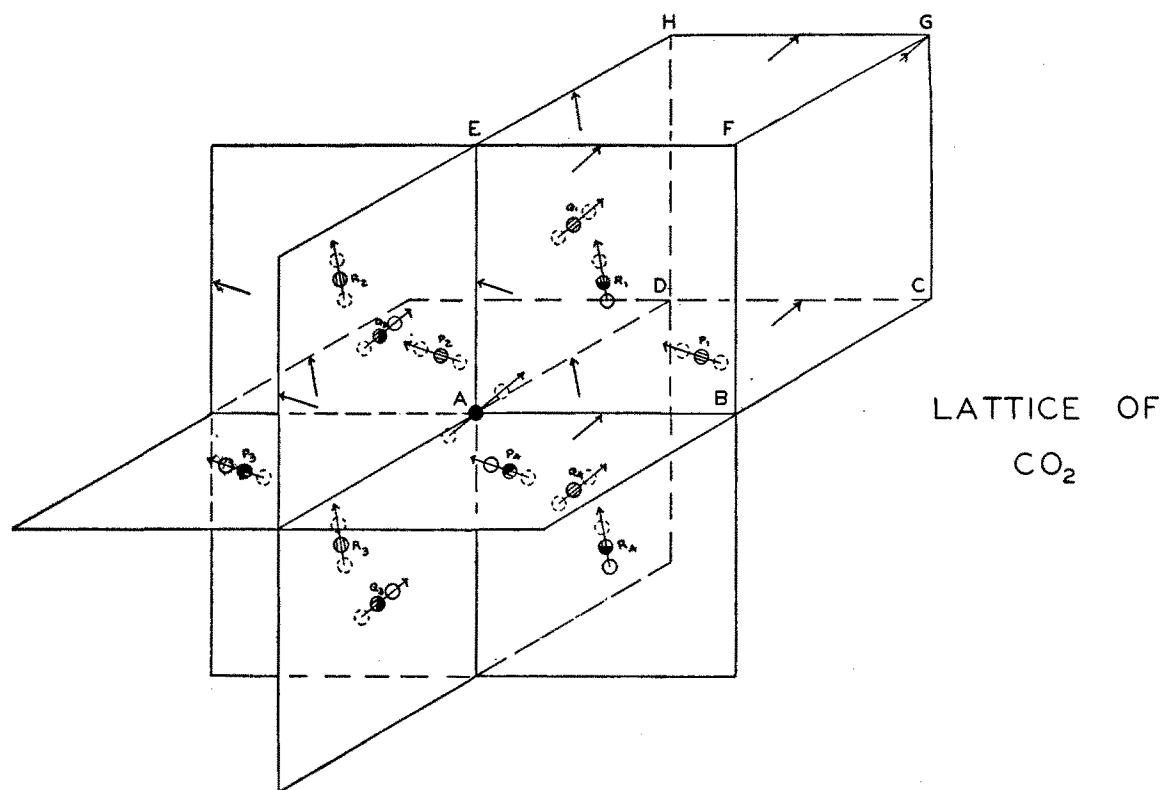[4] See for references, H. A. Stuart, *Molekülstruktur* (1935), p. 185.

FIG. 1. Arrows show directions of different carbon dioxide molecules. Parallel molecules in same plane are denoted with same letter.

the anisotropy of the polarizability which is 2 : 1 in the case of CO_2. Hence we obtain from Silberstein's theory $r = 1.76A$ for the distance between the two polarizabilities and $\alpha = 1.17 \cdot 10^{-24}$ for their value. With the described model the distances from the two centers of one fixed molecule A to those of the surrounding 12 molecules in the face centers and to the 6 closest corners of the cubes are shown in the following illustrations.[5] The summation which has been extended from the shortest distance to $r = a = 5.54A$ gives $\sum(1/R_i^6) = 2.74 \cdot 10^{46}$ cm^{-6}; the integral over the rest, multiplied by the number of centers of attraction per cm^3, gives $0.12 \cdot 10^{46}$ cm^{-6}. If we now use for the characteristic quantity $h\nu$ in formula (1) the ionization energy[6] of the CO_2 molecule $I = 14.4$ ev under the assumption that

this is also justified in our two-center model, we obtain a lattice energy of 5.0 kcal./mole whereas the experimental value is 6.44 kcal./ mole.[7] One may obtain a figure higher than 5.0 kcal./mole if the specific frequency ν in formula (1) is taken from dispersion data rather than from the ionization potential. The dispersion formula to be used in connection with formula (1) should consist of a single term only. Such a formula has been given by Cuthbertson[8] and has been replaced by Fuchs[9] by a detailed formula containing several terms. We have taken 15.5 ev as characteristic energy for a one term formula and inserting it in London's equation have calculated 5.3 kcal./mole for the lattice energy.

How much we are in error by using the frequencies of the CO_2 molecule for the two

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[5] There are 2 molecules with the relative position of A to Q_1, 2 with the relative position of A to P_1, 2 with that of A to Q_2, 2 with that of A to P_2, 4 with that of A to R_1, 6 with that of A to B. The shortest distance between two centers is 2.485A.

[6] This value has been taken from electron impact measurements and hence is to be considered as an upper limit.

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[7] Cf. Landolt-Börnstein, *Tabellen*, III. Erg. Band, 3. Teil, p. 2715.

[8] C. and M. Cuthbertson, Proc. Roy. Soc. Lond. **A97**, 152 (1920).

[9] O. Fuchs, Zeits. f. Physik **46**, 519 (1927).

centers of our model cannot be estimated, but we think that the error will not be large because the CO and CO_2 molecules have almost the same ionization potentials and have absorption spectra in the same far ultraviolet region. We have furthermore used London's dispersion formula in connection with Fuchs' dispersion equation for CO_2.[10] Here no α value need be known, the dispersion data being sufficient to build up the attractive force. But we neglect in this case the existence of an anisotropy of the polarizability which will make our calculated value larger than it actually is. Since the knowledge of α is not necessary we have not used here Silberstein's theory at all but have estimated the positions of the two centers in the following rather arbitrary way. We located namely the center of gravity of the negative charges in the same way as L. Meyer[11] has located the binding moment. This is not unreasonable, as the electrons which contribute to the binding moment are part of the dispersion electrons. Thus we placed the centers $\frac{3}{4}f = 0.84A$ apart from the C atom. Calculation gives now 6.6 kcal./mole for the lattice energy.
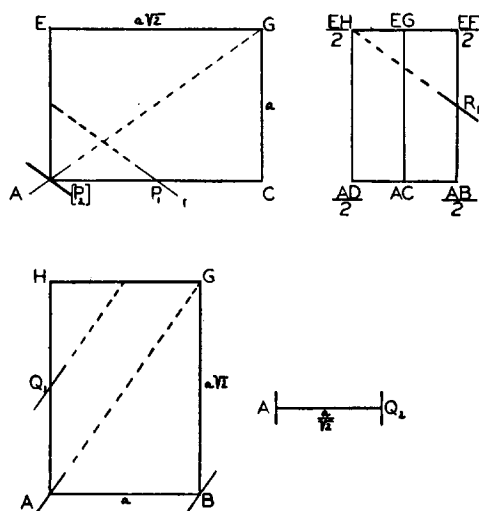

FIG. 2. The brackets indicate that P_2 lies $a/\sqrt{2}$ above the plane of drawing.

[10] The dispersion equation of Fuchs is based upon empirical data in the infrared and in the region from 6000 to 2000A. But since the first principal term contains 720.4A as wave-length of the main frequency, the extrapolation seems rather wide. The observed absorption in the region of 720A is strong and mainly discontinuous.

[11] L. Meyer, Zeits. f. physik. Chemie B8, 27 (1930).

Finally we have carried through the calculation of the lattice energy with a location of the two centers of attraction as in the case of the dispersion formula, but using again formula (1) and at the same time half of the mean value of α for each center, i.e., $\alpha/2 = 13.25 \cdot 10^{-25}$. This model gives a lattice energy of 5.9 kcal./mole when using the ionization potential of 14.4 ev and 6.3 kcal./mole when using $h\nu = 15.5$ ev. Both values would be lowered if the anisotropy of the polarizability would be taken into account.

Summarizing we see that the lattice energies for our model lie between 5.0 and 6.6 kcal./mole with probably the best value of 5.3 kcal./mole.

SECOND APPROXIMATION OF THE ATTRACTION ENERGY

For the second approximation Margenau[12] has given a formula of the dipole-dipole and dipole-quadrupole interaction. The formula contains mean values of powers of the distance of the electrons from the nucleus. Expressing them with the help of the polarizability and the characteristic energy and neglecting thereby the different ways in which the average was taken, we can write the total attraction energy in the form:

$$\Phi = -\tfrac{3}{4}h\nu\alpha^2\frac{N_L}{2}\left(\sum_i \frac{1}{R_i^6} + \frac{9\alpha h\nu}{2e^2}\sum_i \frac{1}{R_i^8}\right), \quad (2)$$

where e is the elementary charge. The results for our different models are shown in Table I; the second term amounts to 52–56 percent of the first. (In Margenau's calculation for hydrogen and helium where he used the actual distribution function of the electrons instead of introducing α, the second term amounted to 25–50 percent of the first.)

OTHER METHODS OF CALCULATING THE ATTRACTION ENERGY

For comparison, we have also used a formula derived by Slater and Kirkwood[13] with a different method:

$$\Phi = -163\cdot 10^{-12}n^{\frac{1}{2}}\alpha^{\frac{3}{2}}\frac{N_L}{2}\sum_i \frac{1}{R_i^6}, \quad (3)$$

[12] H. Margenau, Phys. Rev. 38, 747 (1931).
[13] J. C. Slater and J. G. Kirkwood, Phys. Rev. 37, 682 (1931).

where n is the number of electrons in the outer shell (here $n=8$ for each of the two centers), the other letters having the same meanings as noted earlier. It is very difficult to estimate how reliable this method is, as already London[14] has pointed out. The results are included in Table I. They are of the same order of magnitude as those obtained with London's formula including the dipole-quadrupole interaction (although they do not contain this term, which should still be added[15]) and much higher than the experimental value. A still larger value is obtained when using Kirkwood's[16] formula. This can be understood from the fact that this formula can approximately be reduced to the above-mentioned Eq. (3) with the only change that n then means the total number of electrons in the system considered. Hellmann's method[17] would give a value lying between those obtained with the last two methods, for he takes all electrons into consideration, but with a different parameter for each shell and taking into account the Pauli principle.

We have not used these values in further considerations because they are still larger than the one obtained from Slater and Kirkwood's method.

ELECTROSTATIC ENERGY

CO_2 has two dipoles compensating each other. Their moment has been estimated by Smallwood[18] from infrared dispersion data as $\mu = 1.2 \cdot 10^{-18}$ e.s.u., by Lenel[3] from measurements of adsorption heats of CO_2 on crystals of KCl and KJ

being $\mu = 1.8 \cdot 10^{-18}$ e.s.u. Smallwood's method is in principle only capable to determine the change of μ with vibration; Lenel's method, although correct in principle, is based upon rough estimation which can only give the order of magnitude. Hence both figures do not represent final values, but they very likely give the right order of magnitude. In order to calculate the electrostatic energy terms connected with these dipoles, one has to make an assumption about their position and their length. Adopting the usual picture of dipoles as point charges with negligible distances and locating them at $f=0.84$A as would follow from Meyer's conceptions,[19] we have applied the ordinary dipole formula[20] $(\mu^2/R^3)(-2 \cos \theta_1 \cos \theta_2 + \sin \theta_1 \sin \theta_2 \cos \varphi)$ with the result that a repulsion of $\mu^2 \cdot 1.52 \cdot 10^{36}$ kcal./mole should be present. Inserting $\mu = 1.8 \cdot 10^{-18}$ a value of 4.9 kcal./mole results, whereas with $\mu = 1.2 \cdot 10^{-18}$ e.s.u. 2.2 kcal./mole are obtained. The first value seems impossible and the second improbable. The results indicate that the distances within our lattice are not large enough to justify the application of the dipole formula. As soon as we locate the dipoles halfway between the C and O atoms on both sides and thus increase the distances between the dipoles in different molecules we obtain the much smaller values of 1.3 and 0.6 kcal./mole, respectively (corresponding to $\mu^2 \cdot 0.4 \cdot 10^{36}$ kcal./mole).

But we believe that the charge distribution is much better described with a model of point charges than with dipoles. We have represented it in a very simplified way by means of expressions like

$$\frac{N_L}{2}\epsilon \sum_i \frac{\epsilon_i}{R_i},$$

[14] F. London, Trans. Faraday Soc. 33, 8 (1937), especially p. 16 and 44.

[15] In an abstract Ch. H. Page (Phys. Rev. 51, 1002 (1937)) has very recently calculated the second approximation for the Slater-Kirkwood formula in the case of He and finds an increase of 13 percent when compared to Margenau's 26 percent in this special case; but, as he added when presenting the paper at the Meeting of the Am. Phys. Soc. in Washington, Apr. 29, 1937, the dipole-dipole term becomes simultaneously smaller when the series is expanded. The result is then that the total energy is increased only by 1 percent. If this result should be capable of a generalization and hence could be applied to our case, the result of the variation method would be the same as that of London's method including the second approximation.

[16] J. G. Kirkwood, Physik. Zeits. 33, 57 (1932); A. Müller, Proc. Roy. Soc. Lond. A154, 624 (1936).

[17] H. Hellmann, Acta physicochim. U.S.S.R. 2, 273 (1935); 1, 913 (1935); J. Chem. Phys. 3, 61 (1935).

[18] H. M. Smallwood, Phys. Rev. 41, 164 (1932).

[19] Different authors combine different conceptions with a given dipole moment. Taking, for instance, the dipole moment of the $C-Cl$ bond, $\mu = 1.5 \cdot 10^{-18}$, L. Meyer pictures it as point charges with negligible distance situated at $f = 1.62$A $= \frac{1}{8}$ of the distance $C-Cl$. H. M. Smallwood and K. F. Herzfeld (J. Am. Chem. Soc. 52, 1923 (1930)) also described it as point charges with negligible distance, but situated at $f = 0.77$A, the radius of the C atom, whereas G. C. Hampson and A. Weissberger (J. Am. Chem. Soc. 58, 2111 (1936)) picture it as consisting of two point charges located in the centers of the C and Cl atoms. The difference in results obtained with the different models depends of course more or less on the special problem.

[20] Where θ_i are the angles between the dipoles and their distance from each other and φ the angle around the distance as axis.

where ϵ is the charge of the "dipole." We realize that it is rather incorrect to use here still the word dipole for charges which are quite a distance apart from each other, but it seems a general custom. We have taken the location of the polarizabilities as position of the negative charges and the C atom as center of the positive charges, this picture being very similar to the one used by Lenel.[3] The summation over the electrostatic terms gives an energy of repulsive nature and of value $\mu^2 \cdot 0.085 \cdot 10^{36}$, when using $f = 0.88A$ and $\mu^2 \cdot 0.15 \cdot 10^{36}$ when using $f = 0.84A$. Combining them with Lenel's value we find 0.2 and 0.5 kcal./mole, respectively, while from combining them with Smallwood's figure we obtain 0.1 and 0.2 kcal./mole. These values are much more plausible than the large figures obtained with the dipole formula since the electrostatic forces should not result in a large repulsion when compared to the other terms which constitute the total energy. Part of the repulsion is of course canceled by an attraction due to influence forces resulting from an interaction between the polarizability of one center and the dipole of another center. But these influence forces do not amount to much since the electric fields produced at the position of one polarizable group by the dipoles of all surrounding CO₂ molecules tend to cancel each other. We have not carried out the actual calculation since the electrostatic forces are already small, but we think that the ratio between electrostatic repulsive and influence forces will be about the same as that given by Lenel from a rather elaborate treatment of the influence forces including the inhomogeneity of the electric field. Here the influence forces are about $\frac{1}{4}$ of the electrostatic forces.

It is perhaps not uninteresting to ask what quadrupole moment of CO₂ would result from our assumptions of the charge distribution. Supposing that the distance from a fixed point is large when compared with the dimensions of the molecule, a quadrupole moment of $2 - 3 \cdot 10^{-26}$ e.s.u. would result.

ENERGY OF REPULSION

The distance by which the molecules in the lattice remain separated from each other is determined by the equilibrium between attrac-

tive and repulsive forces. In the case of CO₂ we have found that the electrostatic forces are of repulsive nature, but they do not represent the principal repulsion. Born and Mayer[21] have expressed the repulsive force which results from the quantum mechanical kinematics of the elementary charges, as an exponential function, so that the mutual energy together with van der Waals attraction forces becomes for our example:

$$\Phi = -\frac{A}{R^6} - \frac{B}{R^8} + f(R) + b\sum e^{-(a_i R)/\rho}, \qquad (4)$$

where the first two terms correspond to the van der Waals attraction, the third to the electrostatic energy including influence forces and the fourth to the repulsion. b and ρ are constants and $a_i R$ the distances written as multiples of a smallest distance R. The constant b can be eliminated from the equilibrium condition which is for temperature 0°K and pressure 0

$$\frac{d\Phi(R_{00})}{dR_{00}} = 0 = \frac{6A}{R^7_{00}} + \frac{8B}{R^9_{00}}$$

$$+ \frac{df(R_{00})}{dR_{00}} - \frac{b}{\rho}\sum_i a_i e^{-(a_i R_{00})/\rho}. \qquad (5)$$

Born and Mayer have determined R/ρ for the alkali halides with the aid of thermodynamical relations in which the compressibility β and the change in volume with temperature at constant pressure are involved. Keesom and Köhler[22] have determined $(1/V)(\partial V/\partial T)_P = 5.07 \cdot 10^{-6} T$ for temperature ranges between 20°K and 114°K, while Maass and Barnes[23] have measured it near the sublimation point and have found it equal to 0.00128 which is larger than a linear extrapolation of Keesom's value would indicate. The compressibility β has been roughly measured by Bridgman[24] after this paper had been pre-

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[21] M. Born and J. E. Mayer, Zeits. f. Physik **75**, 1 (1932).

[22] W. H. Keesom and I. W. L. Köhler, Physica **1**, 655 (1934).

[23] O. Maass and W. H. Barnes, Proc. Roy. Soc. Lond. **A111**, 224 (1926).

[24] We wish to express our sincere thanks to Professor Bridgman that he very kindly made these measurements for us and allowed us to use his results in advance of his own publication. Before we had the value of β we could only compare the calculated attraction energy with the experimental lattice energy and from the difference of the two compute a probable value of β.

sented at the Washington meeting of the American Physical Society. He found $\beta = 2.1 \cdot 10^{-11}$ at the sublimation point $T = 195°K$ as an average value between atmospheric pressure and 2500 kg/cm².[25] Since β is not known for the conditions under which Eq. (5) holds and since an extrapolation of β seems very doubtful, we have used the equilibrium condition for a temperature T and pressure 0 which involves a different equilibrium distance $R_{T,0}$. Born has shown that the lattice energy which can be calculated for these different conditions differs by less than 1 percent. Hence our equilibrium condition is

$$R_{T,0}\frac{d\Phi(R_{T,0})}{dR_{T,0}} = \frac{3VT}{N_L}\left(\frac{\partial P}{\partial T}\right)_V = \frac{3T}{\beta N_L}\left(\frac{\partial V}{\partial T}\right)_P$$

$$= \frac{6A}{R^6_{T,0}} + \frac{8B}{R^8_{T,0}} + R_{T,0}\frac{df(R_{T,0})}{dR_{T,0}}$$

$$- b\frac{R_{T,0}}{\rho}\sum_i a_i e^{-(a_iR_{T,0})/\rho} \quad (6)$$

where T, in our case, is 195°K. Within our limit of precision we may neglect the electrostatic term $f(R_{T,0})$, and furthermore put

$$\sum e^{-(a_iR_{T,0})/\rho} \sim \sum a_i e^{-(a_iR_{T,0})/\rho}$$

and

$$R^2_{T,0}\frac{d^2\Phi(R_{T,0})}{dR^2_{T,0}} = \frac{9V}{N_L\beta},$$

which gives us

$$\frac{R_{T,0}}{\rho} = \frac{\dfrac{7\cdot6A}{R^6_{T,0}} + \dfrac{9\cdot8B}{R^8_{T,0}} + \dfrac{9V}{N_L\beta}}{\dfrac{6A}{R^6_{T,0}} + \dfrac{8B}{R^8_{T,0}} - \dfrac{3T}{N_L\beta}\left(\dfrac{\partial V}{\partial T}\right)_P}.$$

Using $(1/V)(\partial V/\partial T)_P = 0.00128$ we obtain

[25] It is perhaps of interest to compare the values of β and $(1/V)(\partial V/\partial T)$ for the few molecular crystals, for which we could find such data, namely PbCl₂, CdCl₂, and C₆H₆. The data are for PbCl₂: $\beta = 3.4\cdot10^{-12}$ and $(1/V)(\partial V/\partial T) = 9\cdot10^{-5}$; for CdCl₂: $\beta = 5.95\cdot10^{-12}$ and $(1/V)(\partial V/\partial T) = 7.3\cdot10^{-5}$ and for C₆H₆: $\beta = 32.9\cdot10^{-12}$ and $(1/V)(\partial V/\partial T) = 35\cdot10^{-5}$. PbCl₂ is according to H. Braekken (Zeits. f. Krist. A83, 222 (1932)) a molecular lattice, whereas CdCl₂ and benzene are more layer lattices. We calculated the repulsive energy with a Born-Mayer formula in analogy to CO₂ as 1.8 kcal./mole for benzene, while the attractive energy gives according to London and Margenau 8.6 kcal./mole and 11.3 kcal./mole with the Slater-Kirkwood formula (J. H. de Boer, Trans. Faraday Soc. 32, 10 (1936)). The experimental value is 10.67 kcal./mole. There are perhaps some other sources of attraction energy not yet considered, which could be more or less nonadditive forces acting in the plane that determines the layers.

$R_{T,0}/\rho = 25.67$ (extrapolation of Keesom's value of $(1/V)(\partial V/\partial T)_P$ would give $R_{T,0}/\rho = 20.74$) and inserting this in Eq. (6) the repulsion term becomes 1.1 kcal./mole (with the extrapolation from Keesom's value 1.7 kcal./mole). Combining this result with our calculation for the attraction energy we see from Table I that we obtain the right order of magnitude for the lattice energy. In fact the figures obtained when using Silberstein's theory are quite close to the experimental value. Of course, our whole theoretical consideration is not accurate enough to allow a decision between the two values.

On account of the rather rough treatment no attempt has been made to calculate small effects, like the energy at zero temperature. The Keesom effect, will not be of importance, because the CO₂ molecules do not rotate in the crystal.

SUMMARY

The above considerations have yielded the interesting result that London's simple theory of van der Waals forces already gives a fair representation of the sublimation heat of the molecular lattice of solid CO₂, indicating that higher approximations, electrostatic and repulsive forces seem to cancel each other. When taking into account these terms a fair agreement between calculated and experimental sublimation heat is obtained. It must be admitted, however, that this is somewhat fortuitous because some simplified assumptions enter the theory. The two-center model itself is an approximation which we consider, however, as a rather good one. Of course, it should be borne in mind that the identification of the optical frequency of the two centers with the frequency of the CO₂ molecule introduces some uncertainty. Besides, we do not know how reliable our application of Silberstein's theory is. It does not work so very well in the case of N₂O which has a lattice very similar to that of CO₂ and very nearly the same lattice energy, furthermore the same number of electrons, but with an anisotropy of the polarizability 3 : 1. Unfortunately there exists such a large variety of possible assumptions in this case on account of the lack of symmetry that

TABLE I.

METHOD OF CALCULATION	LATTICE ENERGY (KCAL./MOLE)		
One center of attraction/molecule:			
London's formula		3.6	
2nd approx. London+Margenau		6.0	
Formula of Slater and Kirkwood		5.7	
Two centers of attraction/molecule:	$\alpha = 13.25 \cdot 10^{-25}$ (average value) $f = 0.84$A $h\nu = 14.4$	$h\nu = 14.4$	$\alpha = 11.7 \cdot 10^{-25}$ (Silberstein) $f = 0.88$A $= 15.5$ ev
London's formula	5.9	5.0	5.3
2nd approx. London+Margenau	9.2	7.6	8.2
Dispersion formula	6.6		
Formula of Slater and Kirkwood	9.6		8.6
Electrostatic energy			
Model of point charges	-0.2 to -0.5		-0.1 to -0.2
Dipole formula	-2.2 to -4.9		
Repulsion energy		-1.1	
Calculated sublimation heat	7.7	6.4	7.0
Experimental sublimation heat	6.44		

no definite conclusions can be drawn. In the case of CO$_2$, however, it is possible, as we have shown, to account theoretically for the observed value.

Our results obtained with different assumptions and approximations are exhibited in Table I.

We wish to express our thanks to Professor E. Teller of George Washington University for valuable suggestions.

A Thermal Conductivity Method for the Determination of Isotopic Exchanges in the Simpler Gaseous Molecules: Erratum

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ON page 386 the sentence: "Our combination consisted of 5000 ohms . . . etc.," should read: "Our combination consisted of 5000 ohms between points *G* and *H* and 8500 ohms between points *E* and *G*."

The Mercury Sensitized Reactions of Methane, Deuteromethanes and the Hydrogen Isotopes: Errata

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IN Tables XIII and XIV on page 221 the superscript "*a*," indicating the use of a Cl$_2$ −Br$_2$ filter, should have appeared *only* on the reaction included in the first line of each table. The second and third line reactions were run using the acetic acid filter.

In Fig. 6 on page 223 the reaction represented by curve *G* should have been included with the *exchange* reactions not with the condensations as indicated.