

## SPONTANEOUS POLARIZATION OF CONDENSING CARBON MONOXIDE AND OTHER GASES WITH AN ELECTRICAL DIPOLE MOMENT

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The electrical polarization which occurs during condensation of gases with a dipole moment is investigated for eight gases at condensation temperatures between 3 and 100 K. On condensate layers of  $10^{-2}$  cm thickness surface potentials up to 100 V were measured. The sign and the value of the polarization depends on the condensation temperature. By means of resonances with the temperature dependent phonon spectrum this polarization and its temperature dependence can be explained.

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### 1. INTRODUCTION

The condensation of gases on cooled surfaces is widely used as a pumping method for physical experiments and for technical applications. In several publications<sup>1-6</sup> the mechanism of gas condensation or adsorption is investigated with respect to these applications.

In the present paper the electrical polarization which occurs on the condensation of gases with a dipole moment is investigated. Polarization of the condensate was observed for  $^{12}\text{CO}$ ,  $^{13}\text{CO}$ , NO,  $\text{N}_2\text{O}$ ,  $\text{SO}_2$ ,  $\text{NH}_3$ ,  $\text{H}_2\text{O}$  and acetone\* at condensation temperature of  $T < 100$  K and pressures of  $p < 10^{-4}$  torr. Parallel measurements with Ne, Ar,  $\text{N}_2$  and  $\text{CO}_2$  showed no polarization.

### 2. APPARATUS AND EXPERIMENTAL METHOD

The UHV-apparatus shown in Fig. 1 was used for the investigation. The condenser surface (2) is a copper plate which is part of a continuous flow cryostat. The temperature of this surface can be varied continuously between 3 and 300 K<sup>8</sup>. The copper plate is mounted on a covar-pyrex-seal (8) and thus it is electrically isolated ( $R > 10^{13} \Omega$ ). The thermal contact between the copper plate and the heat exchanger (3) is achieved by the gas flow through the cryostat. The cooling head of the cryostat is laterally surrounded by a radiation shield (4)

\* Purity of the gases:

$^{12}\text{CO}$ , NO, Ne, Ar,  $\text{N}_2$ ,  $\text{CO}_2$  (99.999%);

$^{13}\text{CO}$  (99.9%+0.1%  $^{12}\text{CO}$ );  $\text{SO}_2$ ,  $\text{N}_2\text{O}$  (99.9%+0.1%  $\text{H}_2\text{O}$ );  $\text{NH}_3$  (99%+1%  $\text{H}_2\text{O}$ );  $\text{H}_2\text{O}$  (99%+1%  $\text{N}_2$ );  $(\text{CH}_3)_2\text{CO}$  (99.5%).

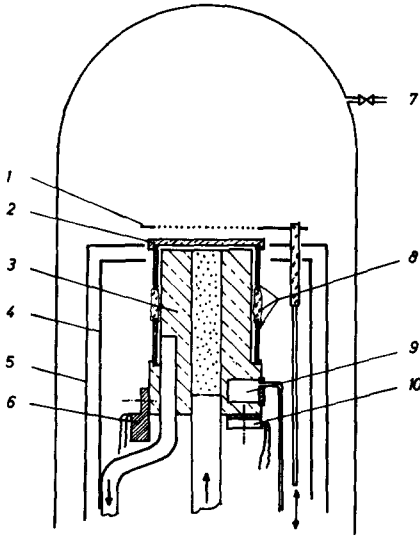


Fig. 1. Apparatus.

cooled by exhaust gas. The cooling head and the radiation shield are surrounded by a further shield (5) kept at room temperature, so that the gas will condense only on the condenser surface (2). Grid (1), which serves as the measuring probe, is located above the condenser surface. The valve (7) serves as the gas inlet.

To measure the charging of the condensate a modified electrostatic potential measuring method is used (Fig. 2). Grid (1) consists of parallel tantalum wires (0.1 mm) which are brazed on to a flat ring of stainless steel (2). Normally the distance between the grid and the condenser surface is 4 mm, but the grid can also be lowered by 1 mm by means of the pneumatic actuator (4).

The thickness of the condensate (3) was always kept below 0.1 mm. By lowering the grid, the capacity between the grid and the surface of the conden-

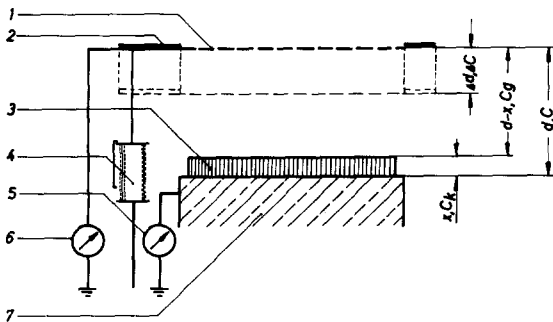


Fig. 2. Electrical measuring system.

sate is raised by  $\Delta C$ . By means of this variable capacity the surface potential  $U$  of the condensate can be measured. The additional charge  $\Delta Q_g$  flowing onto the grid during the lowering is given by

$$U = -\frac{\Delta Q_g}{\Delta C}.$$

It is measured by the electrometer (6), whereas the electrometer (5) shows the charge of the condenser plate. For some measurements the potential of the condenser plate was varied between  $-2000$  and  $+2000$  V.

To explain the experimental method it is necessary first to describe the typical observations:

- (1) During the condensation of gases with a dipole moment the electrometers (5) and (6) indicate currents of  $10^{-14}$  to  $10^{-12}$  A. The currents are equal in size but opposite in sign:

$$I_g = -I_k$$

- (2) The currents are proportional to the rate of condensation  $\dot{M}$  and thus also proportional to the rate of growth of the condensate  $\dot{x}$ :

$$I_g \sim \dot{M} \sim \dot{x}$$

- (3) The surface charging observed is independent of the potential of the condenser plate.

Since the surface charging is independent of electrical fields, it cannot be explained by an accumulation of ionized molecules. Furthermore, as the surface charging was observed only in condensates of gases with a dipole moment, and not in condensates of Ne, Ar,  $N_2$  and  $CO_2$ , it is obvious that it can be explained by polarization, *i.e.* by ordering of the dipole molecules during condensation.

A uniformly polarized condensate consists of layers of equal polarization, *i.e.* equal surface charge (Fig. 3). The surface charge  $Q^*$  directed towards the condenser surface and the free surface charge  $Q$  are equal in size ( $|Q| = |Q^*|$ ). These charges are independent of the condensate thickness  $x$ , but during condensation the free surface charge will be moved towards the grid.  $Q^*$  is compensated by an influence charge on the condenser surface only. However, the free surface charge  $Q$ , which is more distant from the condenser surface, influences charges on the condenser plate ( $Q_k$ ) as well as on the grid ( $Q_g$ ):

$$Q = -(Q_k + Q_g) = \text{const.} \quad (1)$$

The influence charge on the grid is given by<sup>7</sup>

$$Q_g = -Q \frac{x}{\epsilon d - x(\epsilon - 1)}. \quad (2)$$

$\epsilon$  is the dielectric constant of the condensate and  $d$  the distance between the grid and the condenser surface (see Fig. 2). If the thickness of the condensate  $x$  is small compared with the grid distance  $d$ ,  $Q_g$  is proportional to  $x$

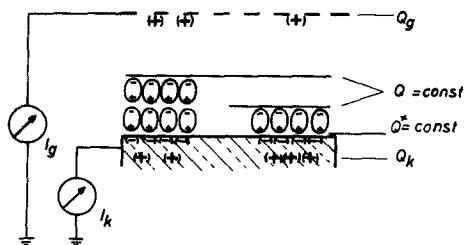


Fig. 3. Model of a uniformly polarized condensate.

$$Q_g = -Q \frac{x}{\epsilon d}. \quad (2a)$$

According to eqn. (1) the currents of influence charge  $I_k$  and  $I_g$  are equal in size and opposite in sign. The current of influence charge on to the grid  $I_g$  follows from eqn. (2a) as

$$I_g = -Q \frac{\dot{x}}{\epsilon d}. \quad (3)$$

Since  $I_g$  is proportional to the rate of growth of the condensate  $\dot{x}$ , it is also proportional to the rate of condensation  $\dot{M}$ :

$$I_g \sim \dot{x} \sim \dot{M}.$$

Thus it is proved that a uniformly polarized condensate will show the typical behaviour mentioned above during condensation.

To investigate the surface charge either the dependence of the surface potential on the condensate thickness, or the current of influence charge  $I_g$  dependent on the rate of condensation  $\dot{M}$  can be measured. The consistency of both methods was repeatedly tested. For observations during condensation, the measurement of the current  $I_g$  is more convenient.

To analyse the results, the ratio of the current of influence charge  $I_g$  to the rate of condensation  $\dot{M}$  is plotted as a function of the temperature of condensation ( $I_g/\dot{M} = f(T)$ ). This method was chosen since  $I_g$  is proportional to  $\dot{M}$  and on the other hand, the ratio  $I_g/\dot{M}$  is proportional to the free surface charge  $Q$ , and therefore also proportional to the average dipole moment  $\bar{\mu}$  of the condensate.

In order to compare the results for the different gases, the ratio of the average dipole moment of the condensate  $\bar{\mu}$  to the dipole moment of the gas molecule has to be calculated. This ratio, which is independent of the dipole moments of the different gases, indicates the degree of polarisation of the condensate. The average dipole moment  $\bar{\mu}$  is the dipole moment of the whole condensate  $Q \cdot x$ , divided by the number of condensed gas molecules  $N$ . According to eqn. (3) it follows:

$$\bar{\mu} = \frac{Q \cdot x}{N} = -I_g \frac{\epsilon d}{\dot{x}} \frac{x}{N}.$$

From  $\dot{M} = nF_k \dot{x} = N\dot{x}/x$  ( $n$ : density of molecules in the condensate) it follows that

$$\bar{\mu} = -\frac{I_g}{M} \varepsilon d \quad (4)$$

With regard to the units\* it follows that

$$\bar{\mu} = -\frac{I_g}{M} \varepsilon d \ 8.6 \times 10^{-11} \text{ c.g.s. cm}, \quad (4a)$$

and the density of the surface charge is given by

$$q = -\frac{I_g}{M} \varepsilon d n \ 2.9 \times 10^{-20} \text{ C/cm}^2 \quad (5)$$

with  $I_g/\dot{M} = [\text{A s/torr l}]$ ,  $d = [\text{cm}]$  and  $n = [\text{molecules/cm}^3]$ .

The characteristic data of the dipole gases investigated are given in Table I (they are taken from ref. 14 if no other source is mentioned).  $\varepsilon$  values of the solid gases were only available at the temperatures indicated, but it can reasonably be assumed that these values are correct, even at lower temperatures.

### 3. RESULTS

The polarization on condensation of the gases  $^{12}\text{CO}$ ,  $^{13}\text{CO}$ ,  $\text{NO}$ ,  $\text{SO}_2$ ,  $(\text{CH}_3)_2\text{CO}$ ,  $\text{H}_2\text{O}$ ,  $\text{N}_2\text{O}$  and  $\text{NH}_3$ , which have dipole moments between 0.1  $D$  and 2.7  $D$  (see Table I), results in surface charges which depend on the temperature of condensation. The sign of the surface charge changes several times depending on the temperature.

By means of the typical results for  $^{12}\text{CO}$  (Fig. 4) the observed phenomena will be discussed. Eight series of measurements with different rates of condensation were conducted. During one series the rate of condensation was held constant and the temperature of condensation was increased or decreased, step by step, starting at 4 to 7 K or 18 to 22 K respectively. Within the limit of accuracy all points are represented by one curve. The deviations can be seen in Fig. 4.

At 4 K a high positive current of influence charge is flowing onto the grid, *i.e.* the condensate has a high negative surface charge. With the increase of temperature the current decreases. At 8.5 K the current disappears, *i.e.* the gas layers which condense at this temperature have no surface charge at all. However, measurements of the potential at this point show that the polarization, and therefore the surface potential of the covered layers, which have been condensed at lower temperatures, has not changed. By raising the temperature further on the nonpolarized layer, a condensate with opposite polarization is formed and so on. There are three further changes of the sign of polarization, *i.e.* the condensate finally consists of a plurality of different polarized layers. At temperatures of 10 and 15.5 K there are maxima of positive surface charge and at 12.5 and 18 K there are maxima of negative surface charge.

\* For the quantity of gas and the rate of condensation the units usual in vacuum technology are used:  
1 torr l =  $5.5 \times 10^{-5}$  Mol =  $3.3 \times 10^{19}$  molecules.

TABLE I

PROPERTIES OF THE GASES WITH A DIPOLE MOMENT INVESTIGATED

Gas	Properties of the molecule				Properties of the crystal					
	M (g/Mol)	$\mu$ (D)*	Structure charge- distribution	$r_1^{**}$ $r_2^{**}$ — ( $\text{\AA}$ )	$\alpha^{**}$ (grad)	$\epsilon$	n ( $10^{22} \text{ cm}^{-3}$ )	L (kcal/Mol)	$Q_D$ (K)	$\omega_D^{***}$ ( $10^{-3} \text{ sec}^{-1}$ )
$^{12}\text{CO}$	28	0.11	C-O	1.13		1.0	2.2	2	$103^{15}$	1.3
$^{13}\text{CO}$	29		- +			(6-60 K)				
NO	30	0.16	N-O	1.3		$1.99^{16}$ (liquid)	3.0	4	$120^{15}$	1.6
$\text{SO}_2$	64	$1.60^{17}$	S O O-	1.43	125	$3.8 (90 \text{ K})^{18}$	1.8	8.5	120	1.6
$(\text{CH}_3)_2\text{CO}$	58	$2.7^{16}$	CO - CH <sub>3</sub> CH <sub>3</sub> + O -					ca. 9		
$\text{H}_2\text{O}$	18	1.84	H H + O -	0.98	105	$3.3^\dagger$ ( $< 80 \text{ K}$ )	3.1	12	192	2.5
$\text{N}_2\text{O}$	44	$0.17^{19}$	N-N-O + -	$r_1 + r_2 = 2.4$	180	$1.99 (100 \text{ K})$	2.14	5.8	120	1.6
$\text{NH}_3$	17	1.46	N - H H H +	$h = 0.4$		$3.4 (90 \text{ K})$	3.0	7.5	215	2.8

Remarks: \*  $1D = 10^{-18} \text{ e g sec cm} = 3.30 \times 10^{-30} \text{ C m}$       \*\*  $r_1 / a$        \*\*\*  $\omega_D = 1.32 \times 10^{11} \theta_D = (\text{sec}^{-1})$

$^\dagger \epsilon = \epsilon_\infty$  at  $T < 80 \text{ K}$  (Ref. 20).

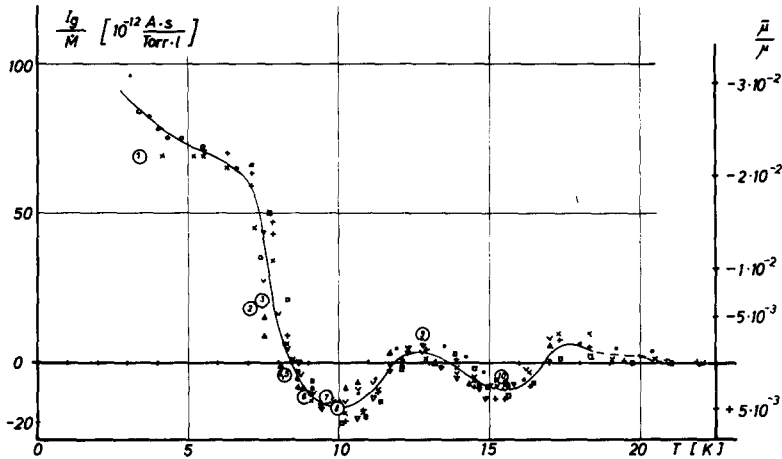


Fig. 4. Polarization of  $^{12}\text{CO}$  as a function of the condensation temperature.

Evidently, the polarization of the condensing layers is not influenced by the properties of the material, on which the condensation takes place. The polarization depends only on the temperature of condensation. This statement is supported by the measurements indicated by numbered circles. Here the gas layer is condensed on to the clean copper plate at a certain temperature, so that a uniformly polarized layer is formed. All resulting values are consistent with the curve. To get further proof that the polarization is independent of the condenser material,  $^{12}\text{CO}$  was condensed at 10 K upon a condensate of water, which had a negative surface charge. The layers of CO immediately showed the positive surface charge given by the temperature of condensation. Therefore the originally negative surface potential decreased with the proceeding condensation of CO. Different materials of the condenser plate, like glass or silver, also had no influence on the results.

The rate of condensation  $\dot{M}$  was varied between  $2.5 \times 10^{-4}$  and  $2 \times 10^{-2}$  torr l/sec. In the same degree the grid current  $I_g$  changes, so that the ratio  $I_g/\dot{M}$  takes up the characteristic value for the condensation temperature. During the measurements, the condenser plate was normally at ground potential. For some tests, however, the potential was varied between  $-2000$  and  $+2000$  V. But even these relatively high fields of  $5 \times 10^3$  V/cm had no noticeable effect on the current of influence charge on to the grid.

For  $^{12}\text{CO}$  the highest temperature at which grid currents were measurable was about 20 K. The surface charge of a polarized condensate produced at a lower temperature and then slowly warmed up, remains in most cases constant up to 14.5 K. At higher temperatures the condensate starts to depolarize, and 18 K the polarization has disappeared. Accordingly the surface charge measured during the condensation at temperatures higher than 14.5 K is not stable, but fades out after the condensation is completed.

The vapour pressure of CO at 20 K is lower than  $10^{-13}$  torr. According to this low vapour pressure, only the  $10^{-4}$ th part of a monolayer vaporizes during

the warming up to 20 K and one hour at this temperature. Thus the condensate depolarizes without any noticeable evaporation. Cooling down of a depolarized condensate does not yield a polarization, and the application of fields of  $10^4$  V/cm during cooling has no influence either.

As shown in Figs. 5 and 6, results similar to those for  $^{12}\text{CO}$  are obtained for  $^{13}\text{CO}$  and the other gases NO,  $\text{SO}_2$ ,  $(\text{CH}_3)_2\text{CO}$ ,  $\text{H}_2\text{O}$ . The absolute value of the ratio  $I_g/\dot{M}$  of all condensates decreases with increasing condensation tempera-

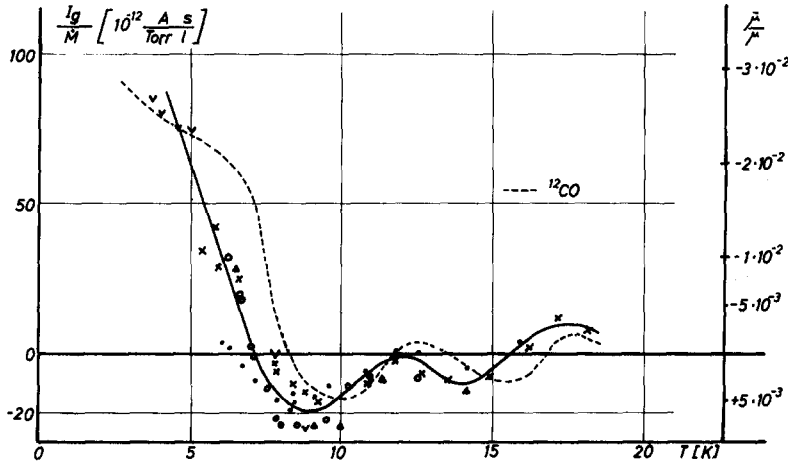


Fig. 5. Polarization of  $^{13}\text{CO}$  and  $^{12}\text{CO}$  as a function of the condensation temperature.

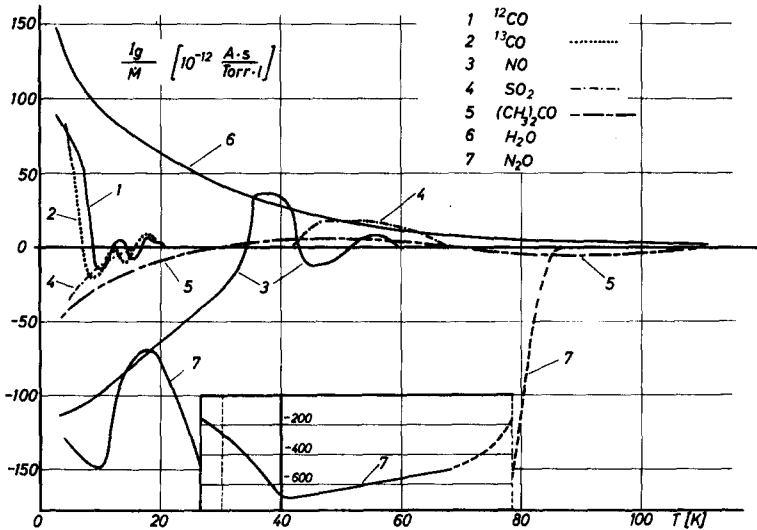


Fig. 6. Polarization of the different gases as a function of the condensation temperature.



ture. For the gases  $^{13}\text{CO}$ ,  $\text{NO}$ ,  $\text{SO}_2$  and  $(\text{CH}_3)_2\text{CO}$  the sign of the polarization changes. On the other hand for  $\text{H}_2\text{O}$  currents only of positive influence charge are observed, *i.e.*  $\text{H}_2\text{O}$  condensates produced below 100 K always have negative surface charges. The vapour pressure of the condensates of  $^{13}\text{CO}$ ,  $\text{NO}$ ,  $\text{SO}_2$ ,  $(\text{CH}_3)_2\text{CO}$  and  $\text{H}_2\text{O}$  is smaller than  $10^{-10}$  torr at the highest temperatures of condensation at which polarizations can be observed. Furthermore for all of these gases the polarization disappears irreversibly when warmed up without evaporation of the condensate.

Layers of  $\text{N}_2\text{O}$ , condensed at temperatures below 30 K, show the same behaviour, but between 30 and 80 K condensates with much higher values of  $I_g/\dot{M}$ , compared with the other gases, are obtained. The polarization of  $\text{N}_2\text{O}$  condensates, formed at temperatures  $>30$  K, is stable in contrast to the polarization at lower temperatures and the polarization of the other gases. This polarization disappears only when the condensate evaporates (between 70 and 90 K the vapour pressure of  $\text{N}_2\text{O}$  increases from  $10^{-9}$  to  $10^{-4}$  torr).

The velocity of depolarization as a function of the temperature was measured for some gases. From the velocity of depolarization the activation energy for the depolarization, *i.e.* the energy which a molecule has to overcome for rotation<sup>7</sup>, can be evaluated. Considering that condensates created at low temperatures consist of small crystals, or are even amorphous, there is good agreement between the activation energy for the depolarization of  $\text{CO}$ , evaluated here, and the activation energy for the rotation of the molecule in the bulk crystal, measured by other authors<sup>9</sup>. For the other gases no comparable values are available.

To estimate the stability of the polarization, the energy of the dipole molecule in the polarized condensates was calculated<sup>7</sup>. In all cases this energy is considerably smaller than the activation energy for depolarization. This shows that polarization is stable until the beginning of thermal depolarization. As the degree of polarization  $\bar{\mu}/\mu$  is proportional to  $I_g/\dot{M}$ , the  $\bar{\mu}/\mu$  scale is inserted in Figs. 4 and 5 as a second ordinate. The sign of this scale was chosen to be identical with the sign of the surface charge. With the exception of  $\text{N}_2\text{O}$  ( $\bar{\mu}/\mu$  up to 0.3), the average dipole moment of the condensate  $\bar{\mu}$  is usually smaller than one percent of the dipole moment of the gas molecule. Despite this small degree of polarization, the surface potential of layers of  $10^{-2}$  cm thickness can be higher than 100 V.

#### 4. HYPOTHESIS OF PHONON RESONANCE

Up to now surface charging during phase transitions has been observed mostly in connection with thunderstorm electricity. The phase boundary causes a separation of charged particles, and the collection of ions results in a surface charge (Costa-Ribeiro effect)<sup>10-12</sup>.

The surface charging effect discussed in this paper cannot be explained by the Costa-Ribeiro effect. Besides the arguments noted above, here a surface charging of condensing  $\text{H}_2\text{O}$  occurs only below 100 K, whereas the Costa-Ribeiro effect causes a charging of  $\text{H}_2\text{O}$  crystals already just below the melting point. Moreover, the surface potentials observed here are much higher. A rather

high surface charging of condensed gases at about 80 K has only been mentioned in a short paper<sup>13</sup>, but no satisfactory explanation has been given.

Since a condensate created at low temperatures consists of small crystals, or is even amorphous, all orientations of the molecules principally have the same probability. The observed surface charging during the condensation of gases with a dipole moment can be explained by a small preference of one special orientation. As the measuring system used here averages over the whole condenser surface, only the components of the dipole moments which are normal to the condenser surface are measured.

The dependence of the polarization on the condensation temperature can be explained by a resonance mechanism between the oscillation frequency of the accommodating molecules and the mean phonon frequency of the condenser material or the condensed gas layer respectively. In the following sections the resonance mechanism will be discussed taking CO as an example.

The CO molecules strike the condenser surface with an energy of rotation and translation according to room temperature. The accommodation takes place within a time of  $10^{-10}$  to  $10^{-11}$  sec. During that time the spare energy has to be stored up in different degrees of freedom. One possibility of movement for a CO molecule on the condensate surface is that of pendulum oscillations, which occur if one end of the molecule (*e.g.* carbon) is fixed to the condensate and the other end (*e.g.* oxygen) can oscillate freely. The potential  $V$  of these oscillations is equal for both possible orientations, whereas the moment of inertia  $I$  depends on the mass of the oscillating atom. Therefore the eigenfrequencies  $\omega_0$  and  $\omega_c$  of the oscillation are determined by the orientation of the molecule and are given by:

$$\omega_0 = \sqrt{\frac{V}{I}} = \sqrt{\frac{V}{r^2 m_0}}; \quad \omega_c = \sqrt{\frac{V}{r^2 m_c}}$$

( $\omega_0$ : oxygen oscillating;  $\omega_c$ : carbon oscillating).

If there is resonance between the mean phonon frequency of the condensate or condenser plate and one of the oscillation frequencies of the accommodating CO molecule, with oxygen at the free oscillating end for instance, then the energy transfer to the condensate is detained for this orientation, as a comparison of the results for  $^{12}\text{CO}$  and  $^{13}\text{CO}$  shows (Fig. 7). The molecules not yet completely accommodated can move on the condensate surface and turn over from one orientation to the other. Therefore, finally, more molecules will be condensed in the nondetained orientation and a negative surface charge will result for the given example.

At low temperatures the dependence of the phonon distribution on temperature and frequency can be described by Debye's approximation. At these temperatures the mean phonon frequency is proportional to the absolute temperature. Therefore, in some temperature regions one orientation is preferred, and in other regions the second orientation. Thus a surface charge of changing sign results, depending on the temperature, as observed in the experiments.

For condensates of the gases with a straight molecule, CO, NO and  $\text{N}_2\text{O}$ , at the lowest temperatures the sign of the surface charge corresponds to that

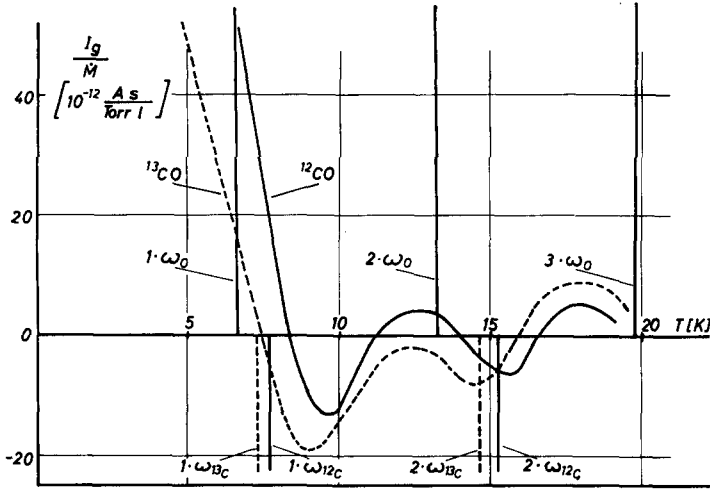


Fig. 7. Comparison of the results for <sup>12</sup>CO and <sup>13</sup>CO with the eigenfrequencies fitted to these results.

orientation of the molecule which has the smaller moment of inertia and thus the higher frequency of oscillation. This is in agreement with the hypothesis of resonance, because at the lowest temperatures the mean phonon frequency is near to the lowest frequency of oscillation and thus the energy transfer for this orientation is hindered. Furthermore the hypothesis of resonance is supported by the fact that the results for <sup>12</sup>CO and <sup>13</sup>CO show characteristic differences, although both gases only differ in their moments of inertia, and not in their chemical properties. Since the potential of the pendulum oscillation at the surface is not known, the oscillation frequencies cannot be calculated. If it is taken for granted that these potentials are equal for <sup>12</sup>CO and <sup>13</sup>CO, the ratio of the eigenfrequencies can be calculated, using the different values for the moment of inertia:

$$\frac{\omega_{12c}}{\omega_0} = 1.15 \quad \text{and} \quad \frac{\omega_{13c}}{\omega_{12c}} = 1.04.$$

Considering these ratios, the eigenfrequencies can be estimated from the results. Figure 8 shows that the maxima of negative surface charge can be explained by the first, second and third resonance with the frequency  $\omega_0 = 1.41 \times 10^{12}$  Hz. The maxima of positive surface charge at about 9 and 15 K correspond in the same way to the frequencies  $\omega_{12c} = 1.62 \times 10^{12}$  Hz and  $\omega_{13c} = 1.55 \times 10^{12}$  Hz. The direction as well as the value of the observed shifting between the curves of <sup>12</sup>CO and <sup>13</sup>CO are in agreement with the hypothesis of resonance.

The average temperature differences between the maxima of the surface charge are proportional to the average eigenfrequency of the pendulum oscillations of the accommodating molecules, i.e.  $\Delta T \sim \omega = \sqrt{V/I}$ . The potentials  $V$  of these oscillations are not known. However, assuming that these potentials are

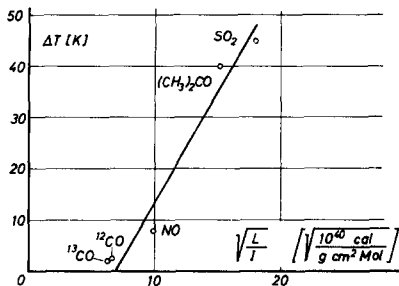


Fig. 8. Comparison of the temperature differences between the maxima of the surface charge of the different gases.

proportional to the total mutual-potential energy of a gas molecule and the condensate, *i.e.* the heat of vaporization  $L$ , it follows:

$$\Delta T \sim \sqrt{\frac{L}{I}}$$

In Fig. 8 these values are plotted for the gases investigated. The diagram shows the expected increase of the temperature difference with increasing moment of inertia. From the extrapolation of the curve for  $\text{H}_2\text{O}$  a hypothetical temperature difference of 200 K results. This explains that the sign of polarization of  $\text{H}_2\text{O}$ , which is stable only below 100 K, does not change.

Since the energy of the observed polarization is small, the rate of condensation as well as the vapour pressure of the condensate are not affected. However, the high surface potential appearing even in very thin condensate layers<sup>13</sup> can disturb electrical systems at low temperatures.

Hence, it can be stated that the polarization of condensing dipole gases, with the exception of some results for  $\text{N}_2\text{O}$ , can be explained very well by a resonance between the mean phonon frequency of the condensate and the oscillation frequency of the accommodating molecules. The results show directly the temperature dependence of the mean phonon frequency at low temperatures. Further information could be obtained by condensation experiments onto a phonon source, with a spectrum which is independent of temperature.

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