

Variation of static permittivity of deuterated ammonium in a strong resonance field

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A theory is developed for the variation of the static permittivity of a relaxing two-level system under saturation conditions. The dependence of the static permittivity of ND_3 on the gas pressure under conditions of saturation of the inversion spectrum by a strong microwave field is investigated experimentally. The relaxation characteristics of the effect are measured for ND_3 by a pulse technique. It is shown that up to a microwave energy flux of 20 kW/cm^2 the variation of the static permittivity is not due to heating of the gas but to changes in the equilibrium distribution of the population of the inversion levels. The relaxation time of the effect in this case is of the same order of magnitude as the rotational relaxation time in the gas.

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1. INTRODUCTION

In the resonance interaction of a strong electromagnetic field with matter, when the frequency of the field is close to the transition frequency for this material, a disruption of the equilibrium distribution of particles over the levels takes place and a number of phenomena are observed as a consequence. One of these phenomena, which was first observed in microwave spectroscopy for the inversion transition in ammonia,^[1] consists in the saturation of the transition under the action of the strong resonance field. Another phenomenon is the change in the absorption coefficient of the material for transitions adjacent to any of the levels of the saturable transition. This phenomenon has been given the name of double resonance and was first used in NMR experiments^[2] and then in gas radiospectroscopy.^[3] Finally, the change in the static magnetic and electric susceptibilities of matter can be observed under saturation conditions. The change in the static magnetic susceptibility of a solid in a strong radiofrequency field was observed in experiments on ferromagnetic resonance.^[4] The observation of a change in the static electric susceptibility for saturation of strong lines of the inversion spectrum of ammonia was reported in^[5], where it was established that the dependence of the change in the static permittivity in a strong resonance field on the frequency of this field is close to the Lorentzian shape of the absorption line and the magnitude of the change reaches a maximum at some optimal pressure. Upon saturation of the ammonium line $J, K=3, 3$, located near 24 GHz, a change in the static permittivity, equal to 10^{-9} , was recorded by microwave radiation with a flux density of 1 mW/cm^2 . Because of the low power of the klystron used, the change was observed only for sufficiently strong absorption lines and with the help of a modulation method.

Meanwhile, it follows from the expression for the change in the static permittivity^{[5,1)}

$$\Delta \epsilon_{\text{max}} = \frac{2}{\sqrt{3}} \frac{\alpha_{\text{max}} \lambda}{4\pi} \frac{\mathcal{E}_0 |P_{mn}|}{\omega_{mn} \hbar} \quad (1)$$

that saturation of the lower-frequency transitions and

use of more powerful sources of microwave radiation should lead to a larger change in the static permittivity than in the case of inversion of the ammonia spectrum (α_{max} is the absorption coefficient at the maximum of the line, λ the wavelength of the radiation, \mathcal{E}_0 the field amplitude). For sufficiently large changes $\Delta \epsilon$, it should be possible to measure the dependence of $\Delta \epsilon$ on the gas pressure and, by use of the pulse method, to determine the relaxation characteristics of the effect.

In this sense, a convenient object of study is deuterated ammonia, the inversion spectrum of which is close to 1.6 GHz. With the use of a source of microwave radiation with a power of 1 W, we can expect an amplification of the effect by almost two orders of magnitude. The example of deuterated ammonia is also of interest since overlap of the absorption lines of the inversion spectrum with the transition from resonance to non-resonance absorption—the so-called collapse phenomenon—is observed for it for the case of relatively low pressures (above 3 Torr).^[6] Under conditions of overlap of the lines, we can also expect a large change in the static permittivity of the gas upon saturation of the inversion transition under the action of the strong resonance field.

The purpose of the present work is the development of the theory of change of the static permittivity of a relaxing two-level system under saturation conditions, the experimental determination of the dependence of the value of the effect on the gas pressure under conditions of partial overlap of the absorption lines of the inversion spectrum of ND_3 , and measurement of the relaxation characteristics of the effect for ND_3 by the pulse method.

In the study of phenomena in which saturation exists by means of powerful sources of radiation, the question always arises as to whether the measured value of the effect is not connected simply with heating of the material. In this connection, estimates are given in the Appendix of the change in the static permittivity from the increase in the temperature of the gas, and also the times of thermal relaxation for ND_3 under the described experimental condition.

2. THEORY

The interaction of a two-level system with an electric field, which is described classically, can be represented by means of the well-known equations of the polarization and the difference in the population of the two-level system^[7]:

$$\begin{aligned} \dot{P} + 2\tau^{-1}P + \omega_0^2 P &= 2\omega_0 \frac{|\mu|^2}{3\hbar} NE, \\ \dot{N} + \tau^{-1}(N - N^e) &= -\frac{2}{\hbar\omega_0} PE. \end{aligned} \quad (2)$$

Here P is the polarization of the system, E the electric field, N the difference in the populations per unit volume, N^e the equilibrium value of the difference in populations per unit volume, ω_0 the angular frequency of the transition, τ the relaxation time of the system, and μ the matrix element of the dipole moment of the transition. In writing down the equations, we have made the following assumptions: the times of longitudinal and transverse relaxation are equal; the correction to the Lorentz coefficient should be equal to unity; P and E are regarded as scalar quantities. All these assumptions are valid under the conditions of the experiment described below.

In the solution of the set of equations (2) for P and N , we have taken into account only those terms which correspond to the effect of saturation. We have considered neither the Stark effect in the solutions, nor effects connected with the transformation of the frequency. We have also not considered the reaction of the dynamic properties of the two-level system on the field.

In the experiment, the saturating microwave field was modulated by rectangular pulses with a large off-duty factor; therefore it is appropriate to consider two cases separately: the stationary regime with continuous microwave pumping and a nonstationary regime with abrupt turning-off the microwave pump.

A. Stationary regime

We define the electric field and the polarization in the following fashion:

$$\begin{aligned} E(t) &= E_\omega \cos \omega t + E_0, \\ P(t) &= E_\omega \chi'_\omega \cos \omega t + E_\omega \chi''_\omega \sin \omega t + E_0 \chi_0, \end{aligned}$$

where $E_\omega \cos \omega t$ is the saturating field with frequency ω close to the transition frequency ω_0 , E_0 is the probing constant electric field, χ'_ω and χ''_ω are the real and imaginary parts of the electric susceptibility at the frequency ω , and χ_0 is the static electric susceptibility.

By solving the set of Eqs. (2) under the condition $\omega_0 - \omega \ll \omega_0$, we get the following for the static electric susceptibility

$$\chi_0 = N^e \frac{|\mu|^2}{3\hbar} \frac{2}{\omega_0} \frac{(\omega_0 - \omega)^2 + \tau^{-2}}{(\omega_0 - \omega)^2 + \tau^{-2} + E_\omega^2 |\mu|^2 / 3\hbar^2}. \quad (3)$$

For the change in the static permittivity under the conditions of saturation, Eq. (3) is transformed into

$$\Delta \epsilon = 4\pi N^e \frac{|\mu|^2}{3\hbar} \frac{2}{\omega_0} \frac{E_\omega^2 |\mu|^2 / 3\hbar^2}{(\omega_0 - \omega)^2 + \tau^{-2} + E_\omega^2 |\mu|^2 / 3\hbar^2}. \quad (4)$$

It is seen from (4) that the dependence of $\Delta \epsilon$ on the frequency of the saturating field ω is close to the Lorentzian shape of the absorption line, in correspondence with the conclusions of^[5]. If we set $\omega = \omega_0$, and recognize that the absorption coefficient at the maximum of the line is equal to

$$\alpha_{\max} = 4\pi N^e \frac{|\mu|^2}{3\hbar} \frac{\omega_0}{c\tau^{-1}},$$

where c is the velocity of light, then we obtain for the maximum change $\Delta \epsilon$ the expression

$$\Delta \epsilon_{\max} = \frac{2}{\sqrt{3}} \frac{\alpha_{\max} \lambda}{4\pi} \frac{E_0 |\mu|}{\omega_0 \hbar},$$

this is identical with (1) apart from the notation.

The expression obtained for $\Delta \epsilon$ (4) is conveniently represented in the form of an explicit dependence on the gas pressure p for further comparison with the experimental results. For this purpose, we introduce the parameter of line broadening $\Delta \nu_p = 1/2\pi\tau p$, the parameter of field broadening of the line $\Delta \nu_E = E_\omega |\mu| / 3^{1/2} \hbar$, we set $\omega = \omega_0$, and denote the coefficient which depends on the intensity of the absorption line by

$$I = 4\pi \frac{\alpha_{\max} c}{\omega_0^2} \Delta \nu_p.$$

Finally, we get the dependence of $\Delta \epsilon$ on p in the form

$$\Delta \epsilon = I p \frac{(\Delta \nu_E)^2}{(\Delta \nu_p)^2 p^2 + (\Delta \nu_E)^2}, \quad (5)$$

where the introduced parameters are measured in the following units: I in Torr⁻¹, p in Torr, $\Delta \nu_E$ in MHz, $\Delta \nu_p$ in MHz/Torr.

B. Nonstationary regime

We define the electric field and the polarization after turning on the microwave pump in the following fashion

$$E = E_0, \quad P(t) = E_0 \chi_0(t).$$

where E_0 is the probing field and $\chi_0(t)$ is the static electric susceptibility. The solution of the set of Eqs. (2) under the condition $\omega_0 - \omega \ll \omega_0$ is of the form

$$\chi_0(t) = N^e \frac{|\mu|^2}{3\hbar} \frac{2}{\omega_0} \left[1 - \frac{E_\omega^2 |\mu|^2 / 3\hbar^2}{(\omega_0 - \omega)^2 + \tau^{-2} + E_\omega^2 |\mu|^2 / 3\hbar^2} e^{-t/\tau} \right].$$

For the change in the static permittivity we obtain

$$\Delta \epsilon(t) = 4\pi N^e \frac{|\mu|^2}{3\hbar} \frac{2}{\omega_0} \frac{E_\omega^2 |\mu|^2 / 3\hbar^2}{(\omega_0 - \omega)^2 + \tau^{-2} + E_\omega^2 |\mu|^2 / 3\hbar^2} e^{-t/\tau}.$$

The explicit dependence of $\Delta \epsilon(t)$ on the gas pressure p can be represented in the following way:

$$\Delta \epsilon(t) = I p \frac{(\Delta \nu_E)^2}{(\Delta \nu_p)^2 p^2 + (\Delta \nu_E)^2} e^{-t/\tau}. \quad (6)$$

3. EXPERIMENT

All the measurements described above were carried out with deuterated ammonia. The content of the basic

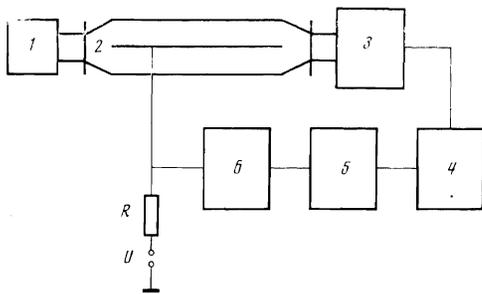


FIG. 1. Scheme of experiment: 1—Apparatus for control of microwave power, 2—measurement capacitor-waveguide, 3—generator of microwave pulses, 4—generator of rectangular pulses, 5—stroboscopic oscilloscope, 6—broadband amplifier.

material in the initial preparation amounted to 99.0%, the air impurity did not exceed 0.1%. Figure 1 shows the experimental setup. The gas was placed in a parallel-plate capacitor formed by the walls of a waveguide with H-shaped cross section and by the central electrode (Fig. 2). The length and the working cross section of the waveguide amounted to 230 cm and 3.2 cm^2 , respectively. The microwave radiation from the generator G4-8 with a power of 1W, modulated by rectangular pulses of length $2 \mu\text{sec}$, with a repetition frequency of 10 kHz, passed through the measurement capacitor-waveguide in the traveling wave mode. The lengths of the fronts of the rectangular microwave pulse did not exceed $0.1 \mu\text{sec}$. To convert the changes of the capacitance of the capacitor-waveguide into an alternating voltage, the capacitor was charged through a sufficiently large resistance R to a potential $U=100 \text{ V}$. In the case of a fast change in the capacitance, due to change in the static permittivity of the gas, the voltage on the capacitor changed by an amount

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

The alternating voltage from the capacitor-waveguide was fed to the input of a broadband amplifier with a time constant of $0.1 \mu\text{sec}$. The amplified signal was recorded by a long-persistence stroboscopic oscilloscope S7-8 in a regime with a memory attachment. [8]

The relaxation characteristics of the change in the static permittivity of ND_3 were determined for the measured time delay of the trailing edge of the rectangular response $\Delta \epsilon(t)$ after turning on the microwave pulse.

The gas pressure in the range from 0.1 to 20 Torr was measured with the help of an oil manometer, and in the range below 0.1 Torr with the help of a thermocouple vacuum gauge calibrated for ND_3 .

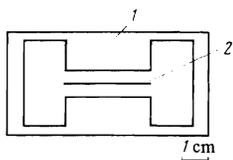


FIG. 2. Cross section of the measurement capacitor-waveguide, 2—central electrode.

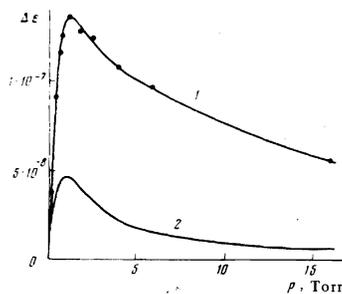


FIG. 3. Dependence of $\Delta \epsilon$ on p for the line $J, K = 3, 3$ of ND_3 , 1—experimental curve, 2—calculated curve.

4. RESULTS OF EXPERIMENT

The typical experimental dependence of $\Delta \epsilon$ on p is shown in Fig. 3. The measurements were carried out with tuning of the microwave generator to the frequency 1615 MHz near the line $J, K = 3, 3$ for ND_3 . The dependence of $\Delta \epsilon$ on p for the single line $J, K = 3, 3$ is shown in this drawing, as calculated from Eq. (5), under the assumption $\Delta \nu_E / \Delta \nu_p = 1 \text{ Torr}$. The value of I for the single line is taken to be $0.95 \times 10^{-7} \text{ Torr}^{-1}$, which, for $\Delta \nu_p = 21 \text{ MHz/Torr}$ corresponds to $\alpha_{\text{max}} = 1.2 \times 10^{-6} \text{ cm}^{-1}$. The form of the experimental dependence of $\Delta \epsilon$ on p is described qualitatively by Eq. (5). However, it is seen that the measured plot of $\Delta \epsilon$ vs. p lies much higher than the calculated curve for the single line. The maximum experimental value of $\Delta \epsilon$ in this case is reached for a pressure of about 1 Torr and is equal to 1.4×10^{-7} , which is approximately three times the maximum value of $\Delta \epsilon$ for the single line. The significant quantitative divergence of the experimental and calculated values of $\Delta \epsilon$ is connected with the effect of the partial overlap of the lines of the inversion spectrum of ND_3 . The overlap of the lines begins to be significant for pressures of about 0.1 Torr and continues to increase with increase in the gas pressure up to 16 Torr.

For a more graphic representation of the transition from the case of a single line to the inversion spectrum of overlapping lines, it is convenient to consider the dependence of $\Delta \epsilon/p$ on p in logarithmic scale. In this case, Eq. (5) takes the form

$$\log \frac{\Delta \epsilon}{p} = \log I + \log \frac{(\Delta \nu_E)^2}{(\Delta \nu_p)^2 p^2 + (\Delta \nu_E)^2}$$

Figure 4 shows such a dependence for tuning of the microwave generator to a frequency of 1627 MHz near the line $J, K = 4, 4$. The drawing also shows two plots calculated for the single line $J, K = 4, 4$ and for the completely overlapping inversion spectrum, under the assumption that $\Delta \nu_E / \Delta \nu_p = 1 \text{ Torr}$. The value of I for the single line, as in Fig. 3, is taken to be $0.95 \times 10^{-7} \text{ Torr}^{-1}$, the value of I in the case of complete overlap of the lines of the inversion spectrum is taken to be $2.86 \times 10^{-6} \text{ Torr}^{-1}$. [9] It is seen in Fig. 4 that as we increase the pressure, the experimental curve 2 rapidly approaches the calculated curve 1. Here complete overlap of the lines of the inversion spectrum of ND_3 occurs for pressures of several tens of Torr. On the low pressure side, even for 0.03 Torr, it appears that the

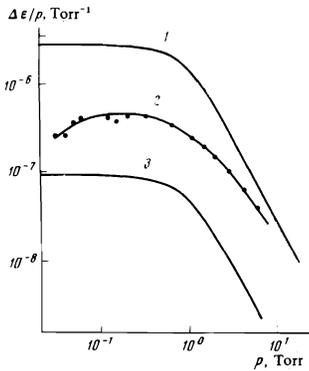


FIG. 4. Dependence of $\Delta\epsilon/p$ on p for the line $J, K = 4, 4$ of ND_3 . 1—calculated curve for $I = 2.86 \times 10^{-6} \text{ Torr}^{-1}$, 2—experimental curve, 3—calculated curve for $I = 0.95 \times 10^{-7} \text{ Torr}^{-1}$.

absorption spectrum of ND_3 cannot be regarded as consisting of nonoverlapping lines.

In both cases—at 1615 MHz and at 1627 MHz—the maximum value of $\Delta\epsilon$ for ND_3 is larger, as was indeed assumed, by two orders of magnitude than the maximum value of $\Delta\epsilon$ for NH_3 .^[5]

Figure 5 shows the results of measurement of the dependence of the relaxation time of the static permittivity of ND_3 on the pressure for tuning of the microwave generator to a frequency of 1627 MHz near the line $J, K = 4, 4$, which was obtained in correspondence with Eq. (6). For gas pressures below 0.05 Torr, the measurements of the relaxation were made difficult because of the smallness of the absolute value of the signal, and over 0.5 Torr, the time delay of the trailing edge of the pulse was too small. The constant relaxation time in the region of pressures from 0.05 Torr to 0.5 Torr is estimated at $21 \pm 3 \text{ MHz/Torr}$. The change in the time delay of the trailing edge of the pulse as a function of the pressure is shown in the photograph, which was obtained from the screen of a stroboscopic oscillograph (Fig. 6).

5. CONCLUSION

From the results that have been given it is seen that the change in the static permittivity of a gas under saturation conditions, as is observed for ND_3 , reaches rather significant values. In our case, the maximum observed change in $\Delta\epsilon$ under saturation conditions, for a pressure of 1 Torr, amounted to 1.4×10^{-7} , which corresponds to a contribution to the static permittivity of approximately three inversion lines of the ND_3 spectrum. Here the total static permittivity of ND_3 for a pressure of 1 Torr is equal to 1.2×10^{-4} ,^[10] and the

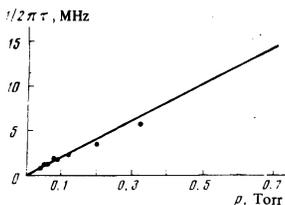


FIG. 5. Dependence of $p\Delta\nu_p$ on p for the line $J, K = 4, 4$ of ND_3 .

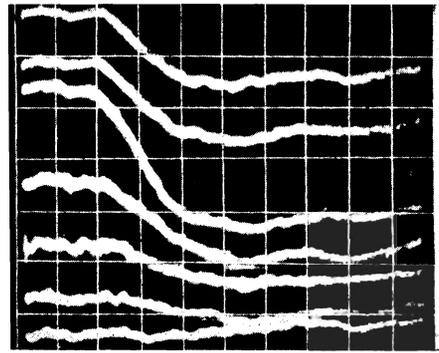


FIG. 6. Trailing edge of a rectangular response $\Delta\epsilon(t)$ for the line $J, K = 4, 4$ of ND_3 for various pressures of ND_3 ; 1—3 Torr, 2—2 Torr, 3—1 Torr, 4—0.5 Torr, 5—0.1 Torr, 6—0.05 Torr, 7—without ND_3 . Time scale: one large division corresponds to 0.1 μsec .

contribution of the entire inversion spectrum to the static permittivity amounts to $\sim 3 \times 10^{-6}$.^[9]

The measured values of the change in the static permittivity $\Delta\epsilon$ and the relaxation time of the static permittivity τ for ND_3 are not connected with the thermal heating of the gas, as estimates show (see the Appendix). The time constant of the thermal relaxation under the conditions of our experiment varied from 10 to 100 μsec , while the relaxation time of the observed effect did not exceed 0.2 μsec over the entire range of pressures used. The change in the static permittivity, due to heating of the gas for an energy flux density of 0.3 W/cm^2 , under the conditions of our experiment, amounted to $\sim 10^{-11}$, while the change in the permittivity due to the observed effect reached 1.4×10^{-7} .

Thus, our results of the study of the change in the static permittivity of ND_3 upon saturation of the lines of the inversion spectrum in a strong resonance field show that, right up to values of the energy flux density of 20 kW/cm^2 , the change in the static permittivity is not associated with heating of the gas, but is due to a change in the equilibrium distribution of populations of the inversion levels, while the relaxation effect is of the same order as the rotational relaxation in the gas.

It is not excluded that the given effect can be observed in more complex multi-level systems, for example, in the saturation of purely rotational or vibrational transitions. Undoubted interest attaches to the study of this effect in the range of higher energy flux densities and higher pressures, right up to the critical values, both in pure gases and in gas mixtures of practical importance.

APPENDIX

We now determine the change $\Delta\epsilon_T$ in the static permittivity as a result of heating in the case of stationary absorption of microwave radiation with energy flux density W . To determine the dependence of $\Delta\epsilon_T$ on W , we start out from the Langevin-Debye formula

$$\epsilon - 1 = 4\pi N(\alpha + \mu^2/kT),$$

where α is the polarizability, μ the dipole moment of

the molecule, and T the gas temperature. For a temperature change ΔT of the gas, we get

$$\Delta \epsilon_T = \frac{\epsilon_{\text{atm}} - 1}{760} p \frac{\mu^2/kT}{\alpha + \mu^2/kT} \frac{\Delta T}{T}, \quad (\text{A. 1})$$

where ϵ_{atm} is the static permittivity of the gas at atmospheric pressure, and p is the gas pressure in Torr.

It now remains to determine ΔT in terms of W with the aid of the stationary heat conduction equation. Taking into account the plane configuration of the capacitor-waveguide used, it is sufficient to consider the one-dimensional heat-conduction equation with a constantly operating heat source:

$$\lambda \frac{d^2 T}{dx^2} + \alpha_{\text{max}} W \eta = 0, \quad \eta = \frac{(\Delta \nu_p)^2 p^2}{(\Delta \nu_p)^2 p^2 + (\Delta \nu_\epsilon)^2}$$

where λ is the coefficient of heat conduction of the gas, α_{max} is the absorption coefficient at the maximum of the line, and the factor η takes into account the saturation of the absorption.

In the case in which the boundary surfaces are located at a distance d from one another and are maintained at room temperature, we have for the maximum change in the gas temperature:

$$\Delta T = \frac{\alpha_{\text{max}} W d^2}{8\lambda} \eta. \quad (\text{A. 2})$$

We introduce a constant that depends on the properties of the gas and the geometry of the apparatus:

$$L = \frac{\epsilon_{\text{atm}} - 1}{760} \frac{\mu^2/kT}{\alpha + \mu^2/kT} \frac{\alpha_{\text{max}} d^2}{8\lambda T} \quad (\text{A. 3})$$

and after substitution of (A. 2) in (A. 1) with account of (A. 3), we obtain

$$\Delta \epsilon_T = L W p \eta. \quad (\text{A. 4})$$

Equation (A. 4) describes the change in the static permittivity of the gas $\Delta \epsilon_T$ at a pressure p , produced by heating or absorption of the UHF radiation with energy flux density W .

If the change $\Delta \epsilon_T$ is regarded at optimal conditions, when $p = \Delta \nu_E / \Delta \nu_p$ and the magnitude of the effect $\Delta \epsilon$ is maximal, according to (5), we then get from (A. 4):

$$\Delta \epsilon_T = L W \Delta \nu_E / 2 \Delta \nu_p, \quad (\text{A. 5})$$

where the parameters are measured in the following units: L in $\text{cm}^2/\text{W-Torr}$, W in W/cm^2 . A similar expression can be obtained for the maximum value of the effect $\Delta \epsilon$ from (5):

$$\Delta \epsilon = I \Delta \nu_E / 2 \Delta \nu_p. \quad (\text{A. 6})$$

We estimate the quantity $\Delta \epsilon_T$ for ND_3 under the conditions of our experiment. For this case, we use the following values of the parameters entering into (A. 3)²⁾:

$$\frac{\epsilon_{\text{atm}} - 1}{760} = 1.13 \cdot 10^{-4} \text{ Torr}^{-1}, \quad \frac{\mu^2/kT}{\alpha + \mu^2/kT} = 0.92, \quad \alpha_{\text{max}} = 1.2 \cdot 10^{-8} \text{ cm}^{-1}, \\ d = 0.5 \text{ cm}, \quad \lambda = 2.47 \cdot 10^{-4} \text{ W/cm-deg}, \quad T = 293 \text{ K}.$$

Substituting these values in (A. 3), we obtain $L = 5.4 \times 10^{-11} \text{ cm}^2/\text{W-Torr}$. Using $W = 3.1 \times 10^{-1} \text{ W}/\text{cm}^2$, $\Delta \nu_E / \Delta \nu_p = 1 \text{ Torr}$ and $I = 0.95 \times 10^{-7} \text{ Torr}^{-1}$, we get for a single line (A. 5) $\Delta \epsilon_T = 8.4 \times 10^{-12}$ and from (A. 6), $\Delta \epsilon = 4.8 \times 10^{-8}$.

It is thus seen that the change in $\Delta \epsilon_T$ associated with heating of the gas in a stationary regime of absorption of microwave radiation is smaller than the values of the effect $\Delta \epsilon$ by more than three orders of magnitude. The dependence of $\Delta \epsilon_T$ and $\Delta \epsilon$ on W is conveniently represented by dividing (A. 5) by (A. 6):

$$\Delta \epsilon_T / \Delta \epsilon = L W / I. \quad (\text{A. 7})$$

From (A. 7), we can estimate the limiting power at which $\Delta \epsilon_T$ becomes equal to $\Delta \epsilon$:

$$W_{\text{lim}} = I / L. \quad (\text{A. 8})$$

In the case of ND_3 , for a single line, the limiting energy flux density W_{lim} amounts to $\sim 20 \text{ kW}/\text{cm}^2$. Under collapse conditions, even for such an energy flux density, the magnitude of the effect $\Delta \epsilon$ will evidently exceed $\Delta \epsilon_T$ by at least an order of magnitude. For $W = 20 \text{ W}/\text{cm}^2$ and optimal pressure, the value of the effect $\Delta \epsilon$ for the single line should amount to $\sim 3 \times 10^{-3}$ and $\sim 10^{-1}$ in the case of complete overlap of the lines of the inversion spectrum of ND_3 .

We now estimate the order of magnitude of the thermal relaxation time τ_T , during which a significant equalization of the temperature in the volume of the gas takes place, after turning on the heat source, when $\Delta \epsilon_T$ tends to zero, and ϵ approaches its equilibrium value at room temperature. For a rough estimate, we can use the formula^[13]

$$\tau_T \sim d^2 / \chi,$$

where χ is the thermal diffusivity coefficient and depends on the gas pressure. Under the conditions of our experiment for ND_3 with the values $\chi = 113 p^{-1} \text{ cm}^2/\text{sec}$, $d = 0.5 \text{ cm}$, we get $\tau_T \sim 2 p \text{ } \mu\text{sec}$. More accurately, the thermal relaxation time can be determined from a solution of the nonstationary equation of heat conduction with a heat source of variable intensity:

$$\frac{1}{\chi} \frac{\partial T}{\partial t} = \frac{d^2 T}{dx^2} + \frac{\alpha_{\text{max}} W}{\lambda} \eta e^{-t/\tau}. \quad (\text{A. 9})$$

$$\tau_T = \frac{1}{\chi} \frac{d^2}{\pi^2}$$

Substituting the data for ND_3 in this formula, we get a somewhat smaller value: $\tau_T \sim 200 p \text{ } \mu\text{sec}$. In the pressure range from 0.05 to 0.5 Torr, which is used in our

experiment for the measurement of the relaxation characteristics of the studied effect, the thermal relaxation time falls in the range from 10 to 100 μsec , which is several orders of magnitude higher than the relaxation time τ of the studied effect.

Thus, we can draw the conclusion that, under the conditions of our experiment, the contribution of the heat change $\Delta\epsilon_T$ and the thermal relaxation τ_T to the measured values of $\Delta\epsilon$ and τ is vanishingly small.

¹The numerical coefficient $2/\sqrt{3}$ was omitted in Eq. (1) of^[5].

²Here and later in the text, the values of the constants for ND_3 are taken from^[11,12]. In those cases in which there are no data for ND_3 the values of the constants for NH_3 are used.

¹C. H. Townes, Phys. Rev. **70**, 665 (1946).

²A. W. Overhauser, Phys. Rev. **92**, 411 (1953).

³N. G. Basov and B. D. Osipov, Opt. spektrosk. **4**, 795 (1958).

⁴N. Bloembergen and R. Damon, Phys. Rev. **85**, 699 (1952).

⁵S. N. Murazin and B. D. Osipov, Opt. Spektrosk. **32**, 430 (1972).

⁶D. R. A. McMahon and I. L. McLaughlin, J. Chem. Phys. **60**, 1966 (1974).

⁷R. H. Pantell and H. E. Puthoff, Fundamentals of Quantum Electronics, Wiley, 1969 (Russ. transl., Mir, 1972, p. 75).

⁸Yu. A. Ryabinin, Stroboskopicheskoe ostsillografirovaniye (Stroboscopic Oscillography) Soviet Radio, 1972, p. 252.

⁹G. Birnbaum and A. A. Maryott, Phys. Rev. **92**, 270 (1953).

¹⁰P. Swarup, Phys. Rev. **104**, 89 (1956).

¹¹Table of Dielectric Constants and Electric Dipole Moments of Substances in the Gaseous State, NBS, Washington, 1953.

¹²S. Chapman and T. G. Cowling, The Mathematical Theory of Non-Uniform Gases, Cambridge Univ. Press, 1970.

¹³L. D. Landau and E. M. Lifshitz, Mekhanika sploshnykh sred (Mechanics of continuous media) Gostekhizdat, 1954, p. 239 [Pergamon, 1958].

¹⁴V. I. Smirnov, Kurs vyssheĭ matematiki (Course of Higher Mathematics) v. IV, Gostekhizdat, 1953, p. 733.

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The analog of Mott scattering of a spin-1 or spin-zero structured particle by a spin-zero target

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The appearance of vector polarization of spin-1 particles, containing two bound particles each of spin 1/2, upon their scattering by spin-zero targets is investigated. A formula is obtained for the polarization under the assumption that in the system of colliding particles there is, in addition to the central field, a relativistic spin-orbit interaction whose operator is proportional to $(\hat{\sigma}_1 + \hat{\sigma}_2) \cdot \mathbf{l}$. It is shown that the polarization tensor, characteristic of the spin correlation, is conserved in the collision, this being a consequence of conservation of the system's total spin in the presence of the spin-orbit interaction. Calculation of the polarization in the system $\text{He}(2^3\text{S}) + \text{He}^{++}$ enables us to predict a maximum polarization $P \sim 0.4$ for the metastable states of helium for energies in the range 1 MeV. Possible experiments with regard to the determination of the polarization and the constants of the optical atomic potential are discussed.

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In 1932, N. Mott predicted the appearance of polarization in a beam of electrons scattered by spin-zero targets.^[1] The reason for the emergence of polarization is the spin-orbit $\mathbf{l} \cdot \mathbf{s}$ interaction, which leads to the appearance of a preferred direction \mathbf{n} of spin orientation. For an initially unpolarized beam this direction coincides with the normal to the scattering plane.

We shall consider the analogous phenomenon of the emergence of polarization as a consequence of the spin-orbit interaction in more complicated systems, which contain two bound particles each with spin $\frac{1}{2}$. Examples of such systems are helium atoms, the deuteron nucleus, etc. In such systems the initial spin state is characterized by a polarization vector \mathbf{P} and by a polarization tensor \hat{Q} . Our goal is the determination of these characteristics after scattering in a system which contains the relativistic $\mathbf{l} \cdot \mathbf{s}$ interaction in addition to a

central field. Below we consider the elastic scattering of particles in triplet ($s=1$) and singlet ($s=0$) states and the possibility of singlet-triplet transitions associated with such collisions.

1. GENERAL CONSIDERATIONS

We shall describe the spin state of a system consisting of two spin- $\frac{1}{2}$ particles by the density matrix ρ . The matrices obtained as direct products of the matrices I , $\sigma_{1\alpha}$, and $\sigma_{2\alpha'}$, given in the spin spaces of the individual particles (I is the identity matrix and $\sigma_{1\alpha}$ and $\sigma_{2\alpha'}$ are the two-dimensional Pauli matrices of the two particles), are introduced as basis matrices. Thus introducing the 4×4 matrices

$$\begin{aligned} I_{(i)} &= I \times I, & \delta_{1\alpha} &= \sigma_{1\alpha} \times I, & \delta_{2\alpha'} &= I \times \sigma_{2\alpha'}, \\ \hat{Q}_{\alpha\alpha'} &= \sigma_{1\alpha} \times \sigma_{2\alpha'} & &= \delta_{1\alpha} \delta_{2\alpha'}, \end{aligned} \quad (1)$$