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PARAMAGNETIC RESONANCE OF FREE GOLD AND SILVER ATOMS TRAPPED IN VARIOUS MEDIA AT LIQUID NITROGEN TEMPERATURES

R. A. ZHITNIKOV and N. V. KOLESNIKOV

A. F. Ioffe Physico-technical Institute, Academy of Sciences, U.S.S.R.

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Trapping of free gold atoms in polar (ordinary and heavy water, ethyl alcohol) and nonpolar (undecane) media is carried out by the condensation method at liquid nitrogen temperatures. The paramagnetic spectra of the trapped atoms are investigated. Paramagnetic resonance of silver atoms trapped in undecane at these same temperatures has also been studied.

THE present research is devoted chiefly to the process of trapping of gold atoms in different matrices when cooled by liquid nitrogen, and to the study of the paramagnetic resonance of the trapped atoms. This work is a continuation of the studies of the trapping and stabilization of free atoms in various media at temperatures close to the temperature of liquid nitrogen, with subsequent study of their paramagnetic resonance spectra. The trapping of silver atoms in nonpolar [1] and polar [2] matrices and a study of the paramagnetic resonance of these atoms were reported earlier.

Part of the present research is also devoted to the study of paramagnetic resonance of silver atoms trapped in undecane.

Specimens containing trapped neutral atoms of gold and silver were obtained by the condensation method on a small-bottom dewar with liquid nitrogen directly in the cavity resonator of a radiospectroscope, as described in [1-3]. The thin small bottom of a quartz dewar, on the external surface of which the sample is located, is cooled from the inner side by liquid nitrogen poured into the dewar. [1-3] Up to now we have assumed that under these conditions a specimen located in a vacuum has the temperature of liquid nitrogen, i.e., 77°K. This temperature had been used by us previously.[1,2] Recently, preliminary measurements carried out by means of a thermocouple attached to the external surface of the small-bottom dewar showed that the temperature there is significantly higher than that of the nitrogen poured into the dewar. Therefore, one can only affirm at the present time that our previous studies,[1,2] and also those described below, were carried out in the region of liquid nitrogen temperature. In several experiments [1,2], the dewar with the nitrogen was pumped on until liquid nitrogen froze on the surface, which obviously also lowered the specimen temperature. Such pumping on is not reflected in any appreciable fashion in the results of the experiments. The working temperatures of our experiments are now under more precise study.

In the present research, a variation of the condensation method is used, developed for matrices having a high vapor pressure at room temperature.[2,3] A previously described [1] 3-cm band radiospectroscope with high-frequency modulation of the magnetic field was used. The accuracy of the measurement of the dc magnetic field was 10^{-4}, and the frequency of the klystron radiospectroscope was determined with an accuracy of 5×10^{-5}. The differential absorption lines are registered by the automatic recorder of the radiospectroscope. The field markers could be placed on the spectrum during the recording process.

GOLD ATOMS IN DIFFERENT MATRICES

Like silver, gold is in the first group of the periodic table and its ground state is $^2S_{1/2}$. The levels of the hyperfine structure of this state in a magnetic field are described by the Breit-Rabi equations:[4,5]

$$W_{I\pm 1/2} = -\frac{\Delta W}{2(2I+1)} + g_I \beta H m \pm \frac{\Delta W}{2}\left(1 + \frac{4m}{2I+1}x + x^2\right)^{1/2}, \tag{1}$$

where ΔW is the energy of the hyperfine splitting in zero magnetic field, I is the spin of the atomic nucleus, $g_I = -\mu_I/\beta I$ is the gyromagnetic ratio for the nucleus, μ_I is the magnetic moment of the nucleus, β is the Bohr magneton, H is the dc magnetic field in which the atom is placed, m is the magnetic quantum number of the total moment of the atom $\mathbf{F} = \mathbf{J} + \mathbf{I}$, where **J** and **I** are the moments of the shell and of the nucleus, respectively, $x = (g_J - g_I)\beta H/\Delta W$ is a dimensionless quantity which is proportional to the magnetic field H, and g_J is the gyromagnetic ratio for the electron shell.

As was pointed out previously,[1] Eq. (1) is applicable not only to absolutely free atoms, but also to atoms trapped in inert media (strictly speaking, this is true for isotropic forces of interaction of the trapped atom with the surrounding medium[7]). The values of ΔW and g_J obtained from Eq. (1) for the trapped atoms will differ somewhat from similar characteristics of the free atom. From a comparison of the values of these quantities for trapped and free atoms, one can decide on the forces of interaction of the trapped atom with the matrix.

The only stable isotope of gold, Au^{197}, has a nuclear spin $I = {}^3\!/_2$ and a positive nuclear magnetic moment. The Zeeman levels of the hyperfine structure of the ground state of gold atoms, calculated from Eq. (1) for $I = {}^3\!/_2$ and $\mu_I > 0$, are shown in Fig. 1 along with the transitions between levels which can be observed in paramagnetic resonance experiments. The form of the paramagnetic resonance spectrum expected for gold atoms is shown below for the three-centimeter range. The Zeeman levels of the free electrons, and the line in the paramagnetic resonance spectrum corresponding to a transition between them with g = 2.00, are shown by dashed lines.

For calculation of the values of ΔW and g_J we can use the two outer lines in the paramagnetic resonance spectrum of gold atoms, corresponding to the transitions (F = 1, m = 1) ⟷ (F = 2, m = 2) and (F = 2, m = −2) ⟷ (F = 2, m = −1). This is convenient, since in this case one has to solve quadratic equations, which can be done easily and with great accuracy. With the other spectral lines it would have been necessary to solve equations of higher degree.

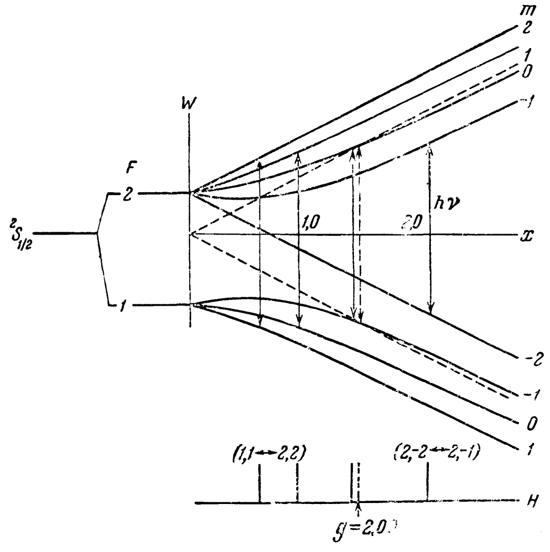

FIG. 1. Energy levels, transitions, and paramagnetic resonance spectrum for the ground state of gold atoms.

The paramagnetic resonance spectra were mapped by varying the magnetic field H at constant frequency ν. With account of this circumstance, Eq. (1) gives the following expressions for the two transitions indicated above:

$$(1,1) \leftrightarrow (2,2),$$

$$\nu = \Delta\nu\left\{\frac{1}{2}(1 + x_1 + x_1^2)^{1/2} + \frac{1}{2}(1 + x_1) + \frac{g_I\beta H_1}{h\Delta\nu}\right\},$$

$$(2,-2) \leftrightarrow (2,-1),$$

$$\nu = \Delta\nu\left\{\frac{1}{2}(1 - x_2 + x_2^2)^{1/2} - \frac{1}{2}(1 - x_2) + \frac{g_I\beta H_2}{h\Delta\nu}\right\}, \tag{2}$$

where $x_1 = (g_J - g_I)\beta H_1/h\Delta\nu$, $x_2 = (g_J - g_I)\beta H_2/h\Delta\nu$; H_1 and H_2 are the values of the magnetic field for the first and second transitions: $\Delta\nu = \Delta W/h$; h is Planck's constant. The nuclear gyromagnetic ratio for gold is $g_I(Au^{197}) = -0.51 \times 10^{-4}$.[5]

Substituting the experimental values of H_1, H_2, and ν for the trapped gold atoms in Eq. (2), one can find $\Delta\nu$ and g_J from this set of equations, these values can then be compared with the similar characteristics of the free atom.

1. Atoms of gold trapped in undecane. The normal paraffin undecane $C_{11}H_{24}$ with a melting point of −25 to −26°C served as the matrix. During the sample preparation the ampoule with undecane [3] had a temperature in the range from 0 to −10°C.

The paramagnetic resonance spectrum of the atoms of gold trapped in undecane is plotted in Fig. 2 (derivatives of the absorption lines are recorded). The spectrum contains four lines, corresponding to four transitions for an atom with nuclear spin $I = {}^3\!/_2$, and is similar to the expected spectrum of gold atoms which is plotted in Fig. 1. In Fig. 2 there is also seen the line with g = 2.00, which is located close to the transition (1, −1) ⟷ (2, 0). This is the line of the free radical formed in the process of trapping of the gold atom in undecane. We did not study the nature of the radical and the mechanism of its formation.

Table I gives the values of $\Delta\nu$ and g_J for gold atoms in undecane, obtained by averaging the results of six experiments, similar to that shown in Fig. 2. The range of errors in the table represents the maximum deviations from the mean values in

Table I

Atom	Matrix	$\Delta\nu$, Mc	$\delta(\Delta\nu)/\Delta\nu$, %	g_J
Au^{197}	Free	6107.1±1.0		2.00412±0.00012
	$C_{11}H_{24}$	5909.0±5.7	—3.24	2.00422±0.00060
	H_2O	6105.6±3.2	—0.02	2.00306±0.00150
	D_2O	6114.4±3.4	+0.12	2.00406±0.00050
	C_2H_5OH	4146.7±13.1	—32.10	2.07150±0.00100
Ag^{107}	Free	1712.56±0.04		2.00224±0.00020
Ag^{109}	"	1976.94±0.04		2.00224±0.00020
Ag^{107}	$C_{11}H_{24}$	1739.9±1.3	+1.60	2.00230±0.00090
Ag^{109}	$C_{11}H_{24}$	2007.3±2.3	+1.54	2.00231±0.00060

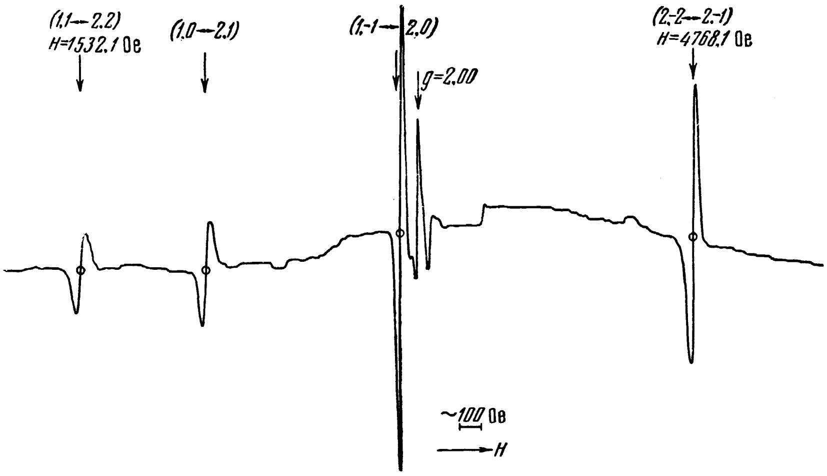

FIG. 2. Paramagnetic resonance spectrum of gold atoms trapped in undecane (ν = 9539.8 Mc)

the series of measurements from which these mean values were determined. In our experiments, these errors always exceeded the instrumental error in field and frequency measurements. The values of $\Delta\nu$ and g_J for free atoms of gold are also shown in the table. These were taken from [5]. For comparison of the values of $\Delta\nu$ of trapped and free atoms, we tabulated the relative shifts of the values of $\Delta\nu$ i.e., $\delta(\Delta\nu)/\Delta\nu$, where $\delta(\Delta\nu) = \Delta\nu - \Delta\nu_{\text{free}}$. For an atom of gold in undecane, the relative shift of $\Delta\nu$ was shown to be equal to −3.2 per cent. It is curious that for silver atoms in undecane this shift (as seen from this same table) is equal to +1.6 per cent, i.e., of sign opposite to the shift in gold.

The intensity of the lines in the spectrum of gold atoms in Fig. 2 is very different for the different transitions, which at first glance seems strange. However, it should be kept in mind that here we have recorded the derivatives of the absorption lines, the amplitudes of which are inversely proportional to the squares of the line widths, since the area of all four absorption lines should be identical, and the area of the line is proportional to the product of the maximum value of the derivative and the square of the line width. Thus the amplitude of the derivative of the absorption line depends strongly on its width. Measurements in the spectrum of Fig. 2 show that all the lines of the gold atom have different widths. The line widths, defined as the distances between the maximum values of the derivative, are plotted in Table II. Even in the same experiment, the line widths change somewhat from measurement to measurement. Table II gives the average value of the widths. Calculations show that, for all four lines of the experimental spectrum, the products of the squares of their widths and the amplitude of the derivatives are actually approximately equal to each other. The reasons why the various lines in the spectrum of the gold atom have the same width are not yet completely clear.

2. Atoms of gold trapped in water. The initial material of the matrix was doubly-distilled water. As noted earlier, [2] the method of obtaining the samples ensures additional distillation and purification of the water in its vapor phase in the vacuum. The temperature of the ampoule of water varied in the range −40°– -60°C, which had little effect on the results of the experiments.

The paramagnetic resonance spectrum of the gold atoms in water agrees with the spectrum of Fig. 2, and contains all four characteristic lines of the gold atoms, at approximately the same

Table II. Line widths (in Oe) for Au^{197} in different matrices

Matrix	Transition			
	(1,1) ↔ (2,2)	(1,0) ↔ (2,1)	(1,−1) ↔ (2,0)	(2,−2) ↔ (2,−1)
$C_{11}H_{24}$	46	39	21	30
H_2O	27	21	11	15
D_2O	26	23	11	15
C_2H_5OH	71	41	28	73

fields as in Fig. 2. Next to the lines (1, −1) ⟷ (2, 0) (in the direction of higher fields) is found the spectrum of the free radical which is produced upon capture of the gold atoms in water. This spectrum of the radical possesses a complicated hyperfine structure and had a very high intensity in our experiments. We did not investigate the spectrum of the free radical. In addition to the lines of gold and of the free radical, a broad, complicated spectrum (or spectra) of comparatively low intensity was also obtained; the nature of this (or these) is unknown to us. This broad spectrum extended on both sides from that of the free radical only about 2600 Oe, but did not prevent recording of the lines of the trapped atoms of gold. This spectrum did not generally reach the outer lines of the gold atoms, from which $\Delta\nu$ and g_J are computed.

Experiments on the capture of gold atoms in water give excellent reproducibility. In Table I are shown the values of $\Delta\nu$ and g_J for atoms of gold, found by averaging the results of four experiments. The small value of the error is the result of a small scatter of the results from experiment to experiment.

For gold atoms in water, $\delta(\Delta\nu)/\Delta\nu = (-0.02 \pm 0.05)$ per cent, i.e., the relative shift of $\Delta\nu$ has a very small negative value, which differs from zero by less than one half the error of experiment. This testifies to the almost complete balance of the forces that compress and stretch the shell of the trapped atom. In individual experiments with gold in water, this relative shift oscillated within the limits of error mentioned and was both negative and positive. For silver atoms in water,[2] the relative shift of $\Delta\nu$ is $(+1.41 \pm 0.15)$ per cent, i.e., it is essentially positive.

The intensities of the lines in the spectrum of gold atoms in water are different, and the ratio of the intensities is about the same as shown in Fig. 2. As in the case of undecane, this is connected with the fact that the lines have different widths. The widths of the lines for gold atoms in water, shown in Table II, are approximately half those for gold atoms in undecane, and the behavior of the widths of the various lines in the spectrum is very similar for these two cases. The products of the squares of the widths and the maximum values of the derivatives also turned out to be approximately equal for all these transitions in the spectrum of gold atoms in water.

3. Atoms of gold trapped in heavy water. In order to make clear whether or not a significant contribution to the line width of paramagnetic resonance is made by the interaction of trapped atoms with the nuclear magnetic moments contained in the matrix, it was decided to carry out experiments with trapping of gold atoms in D_2O. This problem arose in connection with the still unclear mechanism of the different broadening of the various lines in a given spectrum of trapped atoms of gold. Since heavy water differs from ordinary water only in the replacement of protons by deuterons, all the differences in the paramagnetic resonance spectra of the atoms for these two matrices should be determined by the decrease in the value of the nuclear magnetic moments belonging to the matrix.

For these experiments, heavy water was used with a content of D_2O not less than 99.98 per cent. As in the case of ordinary water, the ampoule with heavy water used in obtaining the specimens had a temperature of −40 to −60°C.

The paramagnetic resonance spectra of atoms in heavy and ordinary water proved to be completely identical in form; not only the lines of the atoms of gold and the ratio of their intensities were identical, but also the spectra of free radicals, and also the broad low-intensity spectra described above, the nature of which is still unknown.

It is seen in Table II that the widths of the lines in ordinary and heavy water are identical accurate to the variability of these widths. Consequently, the interaction with the nuclear moments of the matrix does not make a significant contribution to the line width of the paramagnetic resonance of the trapped atoms, and cannot be responsible for the different line widths of the various lines.

The values of $\Delta\nu$ and g_J for the gold atoms in heavy water, obtained from four experiments, are

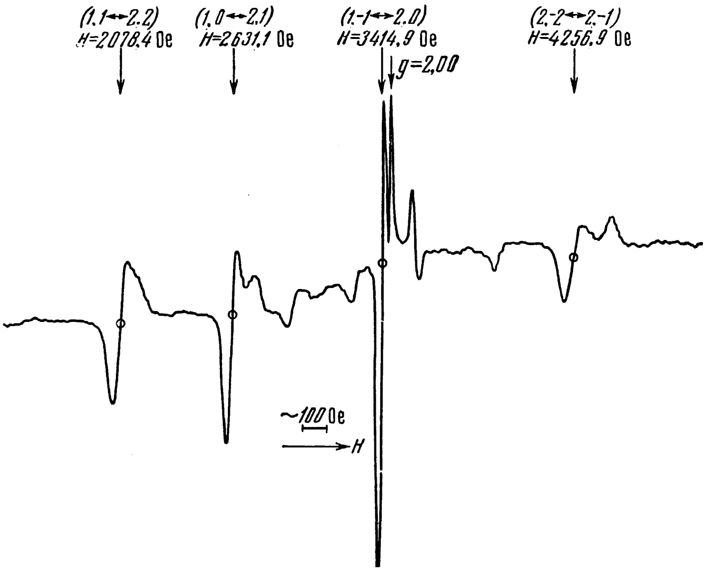

FIG. 3. Paramagnetic resonance spectrum of gold atoms trapped in C_2H_5OH (ν = 9525.3 Mc).

shown in Table I. In this case $\delta(\Delta\nu)/\Delta\nu = (+0.12 \pm 0.05)$ per cent, i.e., the relative shifts of the values of $\Delta\nu$ in heavy and ordinary water are in each case close to each other. The accuracy of our experiments to date is insufficient for a final resolution of the question as to whether there actually exists a small difference between these shifts, or whether they are identically equal.

4. Atoms of gold trapped in ethyl alcohol. In these experiments, absolute ethyl alcohol was used with a purity of 99.5 per cent. In the process of obtaining the specimens, the alcohol was subjected to vacuum distillation and additional purification.[2] The ampoule with the alcohol in this case had a temperature of about −70°C.

Figure 3 shows the paramagnetic resonance spectrum of gold in ethyl alcohol. Four characteristic lines of gold atoms are seen which, however, lie much closer to one another here than in Fig. 2. This testifies to the large negative shift of the quantity $\Delta\nu$ for atoms of gold in alcohol. The lines of gold in Fig. 3 have a different intensity and a different width. The line widths shown in Table II were much larger than in the other matrices; furthermore, a non-proportional large broadening of the high-field $(2, -2) \longleftrightarrow (2, -1)$ line took place.

To the right of the transition $(1, -1) \longleftrightarrow (2, 0)$ in Fig. 3 there is seen a free radical line with g = 2.00. The position of this line coincides with the place of the lines of DPPH. There is also a very broad weak spectrum in Fig. 3, the nature of which is not clear. Additional much weaker (and in certain cases not completely resolved) lines are seen close to the lines of the gold atoms. Most probably, these lines belong to the broad spectrum of unknown origin mentioned above. However, there is some probability that these weak lines belong to atoms of gold trapped in the alcohol at places with an environment of different character than the principal mass of gold atoms.

The reproducibility of the results in experiments with alcohol was quite excellent. Values of $\Delta\nu$ and g_J are given in Table I for gold atoms in alcohol, found from five experiments. The relative shift of the value of $\Delta\nu$ was −32.1 per cent. For silver atoms in ethyl alcohol, this shift was much less, −12.4 per cent.

In all experiments with gold atoms, during the time of the experiments, which extended over several hours, no changes were noted in the paramagnetic resonance spectra, which evidently indicated the excellent stabilization of the trapped gold atoms. Usually, about two per cent of the gold atoms entering the specimen are stabilized in the investigated matrices in the form of isolated atoms that cause the paramagnetic resonance spectrum. For silver, this fraction was about one order of magnitude smaller.

SILVER ATOMS TRAPPED IN UNDECANE

For comparison with gold atoms trapped in undecane, silver atoms trapped in the same matrix were also studied. These same experiments are of interest for a comparison of the behavior of the silver atoms trapped in various paraffins.[1] The ampoule with the undecane had the same temperature as in the experiments with gold, i.e., from 0 to −10°C.

The paramagnetic resonance spectra of the silver atoms in undecane were similar to the spectra obtained earlier [1,2] (they are especially similar to the spectrum of Fig. 1 in [2]) and were calculated in the same way as in [1,2].

The values of $\Delta\nu$ and g_J for silver atoms in undecane, obtained from five experiments, are given in Table I, where similar values are also listed for free silver atoms. The relative shift of the value of $\Delta\nu$ for silver in undecane obtained from these experiments was already used above for a comparison with gold atoms in the same matrix.

CONCLUSION

The results of the present research have shown the possibility of capture and stabilization of isolated atoms of gold in different matrices at the temperature of liquid nitrogen, and also the possibility of observing and studying in detail paramagnetic resonance spectra of trapped atoms. This is the second element, whose atoms can be captured at these temperatures.

The relative shifts of the hyperfine splitting $\delta(\Delta\nu)/\Delta\nu$ for atoms of gold in different matrices, given in Table I, vary in the range from 0.1 to -32.1 per cent. Comparison with the results for silver atoms, which are listed in this table and in the previous researches,[1,2] show that for both elements the change of this shift in the transition from one matrix to another takes place in similar fashion, but for gold the shifts in all matrices are appreciably displaced in the negative direction. The latter evidently indicates that the Van der Waals interaction with the matrix plays a larger role for gold atoms than for silver atoms. For both gold and silver [2] atoms, in such polar media as water and ethyl alcohol, which are similar in their properties, the relative shifts differed very strongly. At the same time, these shifts were rather close in polar water and in nonpolar undecane. The very large negative shift, -32.1 per cent, found for gold in alcohol, is extremely interesting for an atom in the state $^2S_{1/2}$.

The values of $\Delta\nu$ for trapped atoms were obtained from Eq. (1), which is valid in the case of isotropic interaction of an atom with a matrix, i.e., for a symmetrical surrounding of the atom in the matrix. In order to study the degree of anisotropy of this interaction for trapped gold atoms, it would be necessary to compute the values of the quantities $\Delta\nu$ (and also g_J) from various pairs of lines of a given paramagnetic resonance spectrum, and compare them with one another. The divergence of these values, which significantly exceeds the experimental errors, would testify to the necessity of adding an anisotropic term to the Hamiltonian from which Eq. (1) is obtained.[1,7] Such an attempt would be especially interesting for the matrix of ethyl alcohol, where a very large negative shift in the value of $\Delta\nu$ is obtained, and also a large value of g_J. These calculations on the spectra of gold atoms are of interest also for estimating the results obtained on silver atoms, the spectra of which consist entirely of two lines, and to which such a method of procedure is directly inapplicable. Calculations with other line pairs are somewhat more cumbersome than calculations with the outer lines, and we have not yet carried them out for gold atoms.

The values of g_J given in Table I for the trapped atoms differ, with the exception of the case of gold atoms in alcohol, from the values of g_J for free atoms by an amount that does not exceed the experimental errors. For gold atoms in ethyl alcohol, the values of g_J are much larger than for the free atom, and the difference greatly exceeds the experimental error. It is interesting that the value of g_J for atoms of silver in ethyl alcohol is less than for the silver atom.[2]

A theoretical analysis of the properties of trapped atoms of gold and silver, similar to that for hydrogen atoms and alkali metals,[7] is impossible at the present time, since evidence is lacking as to the structure of our matrices, which were obtained by condensation at the temperature of liquid nitrogen, and as to the possible character of the neighboring atoms in these matrices.

As noted above, the differences in the intensities of the various lines in a given paramagnetic resonance spectrum of trapped atoms are attributed to different line widths. The line widths for a gold atom in different matrices are given in Table II. Calculations show that the areas of all four spectral lines of gold agree very well with one another in most cases.

Thus the explanation of the differences between different lines in the spectrum of the gold atoms reduces to an explanation of the reasons for differences in the widths of these lines.

The line width is determined by the following factors: 1) the dipole-dipole interaction, which is characterized by the relaxation time T_2, 2) the spin-lattice relaxations with time T_1, 3) hyperfine interaction of the atom with the nuclear moments of the matrix, 4) the effect of saturation, and 5) the inhomogeneity and anisotropy of the interaction of the atoms with the matrices.

The dipole-dipole interaction obviously does not make a significant contribution to the line width, since different experiments, in which the number and concentration of the trapped gold atoms varied over wide limits, gave identical line widths. The maximum value of the spin-spin relaxation time in our experiments could be estimated in the following way. The number of gold atoms in the specimen, determined by the area of the lines, usually had a value of the order of $2 \times 10^{15} - 2 \times 10^{16}$. Under the assumption of an equilibrium distribution of the atoms over the volume of the specimen, this leads to a concentration of $10^{18} - 10^{18}$ cm^{-3} and to a relaxation time T_2 [6] of about $10^{-6} - 10^{-7}$ sec. This is the upper limit for T_2, since the concentration for a nonequilibrium distribution of the atoms will be larger while the values of T_2 are smaller than that given above.

Experiments with different resonator powers have shown that the saturation effect does not make an important contribution to the line width. Judging from the same experiments it can be assumed that the spin-lattice relaxation is also not decisive at these widths.

In investigations with matrices from ordinary and heavy water, it was shown that replacement of the protons by deuterons does not change the line widths or the form of the spectra.

Since there are essentially only nuclear moments of hydrogen in all matrices used in our researches, these experiments indicate that the hyperfine interaction of the trapped atoms with the nuclear moments of the matrix does not make a considerable contribution to the line widths.

Thus one can evidently assume that the width and shape of the lines of the trapped atoms are determined in practice by the inhomogeneity and anisotropy of the forces of interaction of the atoms with the matrices. Jen et al.[7] have shown that this inhomogeneity of the interaction forces, which can be associated, for example, with some differences in the dimensions of the cavities occupied by various atoms in the matrix, leads in the case of nuclear spin $I = 3/2$ to a larger width of the outer lines of the spectrum in comparison with the inner ones. From Figs. 2 and 3 and Table II, it is seen that the outer lines are actually wider than the inner lines next to them. Moreover, in spectra of gold atoms, the left pair of lines lying in the region of low magnetic field has a greater width than the right pair of lines which is in a high field (the only exception is the outer line in the case of alcohol). The reasons for this phenomenon are not clear. As was mentioned previously,[2] lines from the low field side in the spectra of trapped silver atoms, are also wider, as a rule, than the lines lying in the high field.

It is seen from Table II that the lines of the gold atoms are narrower in ordinary and heavy water and wider in ethyl alcohol. A similar dependence of line width on the matrix takes place also for silver atoms.[1,2] Together with the results of Table I and our previous researches,[1,2] this suggests that, of all the matrices studied, water produces the least perturbation of the shells of the trapped atoms and ethyl alcohol the greatest.

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