

V. MICROWAVE SPECTROSCOPY

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A. INTRODUCTION

Broadly defined, microwave spectroscopy is the study of the interaction of centimeter and millimeter wavelength electromagnetic radiation with matter. The Microwave Spectroscopy group has interested itself in the observation of this interaction and its interpretation in terms of physically significant constants.

Until a year ago, the group concentrated on the interaction of the rotation of molecules in a low-pressure gas with microwave radiation. Some order may be given to the study by classifying molecules as linear, symmetric, or asymmetric, and considering the effect on the absorption spectrum of various internal or external perturbations. These perturbations may be classified, for example, as centrifugal distortion, or nuclear electric quadrupole structure, vibrational structure, isotope structure, and the Stark and Zeeman effects. The Stark and Zeeman effects are used to evaluate molecular electric and magnetic moments.

Some work is continuing with the high-sensitivity spectroscope on the study of vibrational structure. Work is also continuing in the application to specific molecules of the general techniques of analysis previously evolved. The effect of internal rotation is another example of an area of continuing study.

Work is also in progress on paramagnetic spectra in the microwave region. These absorptions exist, primarily, as a result of a static magnetic field which is imposed on the sample. Again, for simplicity, materials studied may be categorized as solids, liquids, or vapors. Work in progress is on solids and a gas (oxygen). The solids are nonconductors and conductors. Here again, the aim is the precise interpretation of the absorption shape, frequency, and amplitude in terms of suitable physical constants or parameters.

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B. PARAMAGNETIC RESONANCE IN OXYGEN GAS

The complete X-band absorption spectrum has been examined by the circular polarization technique described in the Quarterly Progress Report, October 15, 1953. As expected, this has given an unambiguous determination of ΔM for each transition of observable intensity. The results are that there were 95 lines for which $\Delta M = +1$ and 25 lines for which $\Delta M = -1$. All of the strongest lines were of the $\Delta M = +1$ category. This is in agreement with those intensities that have been calculated, as is the fact that $\Delta M = 0$ transitions are too weak to observe. The most intense of the $\Delta M = 0$ lines

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calculated should have an intensity of only about 1 percent of that of the strong $\Delta M = +1$ lines.

Linewidth measurements have been extended, giving the result that $J = K - 1$ levels are perhaps 5 percent broader than the corresponding $J = K + 1$ levels reported in the Quarterly Progress Report, October 15, 1953. Since all the differences are small, higher accuracy is desirable for reliable results. We are preparing to repeat these measurements with a smaller cavity and narrower magnet gap to eliminate the residual width caused by field inhomogeneity over the cavity.

Calculations have been carried out using Meckler's wave function for oxygen (A. Meckler: J. Chem. Phys. 21, 1750, 1953) to evaluate the spin-spin coupling energy contribution to the parameter λ entering into the effective Hamiltonian. The result is only 21 percent of the observed value. It is expected that use of more accurate atomic orbitals than gaussians will increase λ by a factor of at least two. This point is being investigated at the present time.

M. Tinkham

C. PARAMAGNETIC RESONANCE IN METALS

Paramagnetic resonance absorption by conduction electrons in metals at microwave frequencies (9000 Mc/sec) is being investigated. Samples of metallic sodium ultrasonically dispersed in paraffin have been used. Particle size measurements that have been made indicate that the majority of the sodium particles are of the order of magnitude of the skin depth (0.9μ at 9000 Mc/sec). It is desirable to have the particles as small as the skin depth in order to eliminate linewidth broadening from random phase and frequency modulation of the microwave field as seen by the electron.

The observed resonance absorption was, as theoretically expected, independent of temperature and had a linewidth which varied approximately linearly with temperature. The widths of the resonance curve at half-maximum power points were 11.3 gauss at room temperature, 4.0 gauss at 77°K, and 1.8 gauss at 4.2°K. The linewidth does not extrapolate to zero at 0°K. This probably indicates that there are two relaxation mechanisms contributing to the broadening. One is a temperature-dependent mechanism believed to be caused by an interaction of the spin-orbit type; the other is independent of temperature and possibly is caused by the effect of particle size. The spin resonance in sodium, as first observed by Griswold et al. (T. W. Griswold, A. F. Kip, C. Kittel: Phys. Rev. 88, 951, 1952), had a linewidth of 78 gauss and no temperature dependence. Their result probably came from having too large particles.

Precise measurements have been made of the g -value of the absorption. The microwave frequency was measured directly with a frequency standard. The magnetic field was measured by proton resonance. The resulting g -values for sodium are: $g = 2.0012 \pm 0.0002$ at room temperature, $g = 2.0012 \pm 0.0002$ at 77°K, and $g = 2.0014$

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± 0.0002 at 4.2°K . These results disagree with the theoretical result of 2.0019 as calculated by Yafet (Y. Yafet: Phys. Rev. 85, 478, 1952).

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D. PARAMAGNETIC RESONANCE IN AMMONIUM CHROMIUM ALUM

Paramagnetic resonance experiments have been conducted on single crystals of a solid solution (ammonium chromium alum in ammonium aluminum alum). Dilutions of approximately one in six and approximately one in seventy were used.

These data were obtained at X-band:

	Dilution 1:6	Dilution 1:70
Linewidth at half-power	55-85 gauss	30-55 gauss
g 300°K	1.9776 ± 0.0002	1.9781 ± 0.0001
g 77°K	1.9765 ± 0.0002	1.9765 ± 0.0002
g 4°K	1.9766 ± 0.0004	—
δ 300°K	0.099 cm^{-1}	0.088 cm^{-1}
δ 77°K	0.0056 cm^{-1}	—

The error expressed in the g -factor is the maximum deviation of the observations. Linewidth varies strongly from one line of the spectrum to another; the limiting values are expressed here. Data obtained at S-band correlate with these values and theory to 0.3 percent.

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