Letters to the Editor

Paramagnetic Resonance Reorientation of Atoms and Ions Aligned by Electron Impact

H. G. Dehmelt

University of Washington, Seattle, Washington
(Received June 29, 1956)

SIMPLE angular-momentum conservation considerations indicate that considerable alignment should result when atoms are excited to non-S states by electron impact. If, e.g., the energy of the impinging electron is just above that of the level to be excited, it will nearly be at rest after the collision. The angular-momentum transmitted to the atom can then have no component in the direction of the exciting electron beam and, therefore, the selection rule $\Delta M_L = 0$ applies. This is borne out by experiments[1] on the polarization of the light emitted by electron impact excited atoms and also in accordance with a more refined theoretical treatment of the collision process.[2] Similar considerations apply to the formation of ions from neutral atoms and molecules. With ions, the intriguing possibility even exists to trap them by suitable fields[3] and maybe preserve the alignment for milliseconds. The great variety of states that can be reached by electron impact gives this excitation method a marked advantage over the optical excitation used in the related "Double Resonance" experiments.[4]

Once the alignment has been achieved under conditions favorable to its preservation for a sufficiently long time, the angular momenta can be reoriented by paramagnetic resonance, the resonance being the sharper the longer the alignment persists. The central problem of detecting the paramagnetic resonance is thus transformed into that of monitoring the alignment. A variety of ways exist to accomplish this. Besides, as already mentioned, observing the *emitted* optical radiation with respect to polarization and/or directional distribution, it is possible in the case of metastable or long-living states to observe the *absorption* of polarized optical radiation corresponding to a transition to some higher state. A further possibility is based on the angular dependence of the ionization cross section. It is quite possible to collect the ions produced by a parallel electron beam fast enough to ionize from the metastable level but too slow to do so
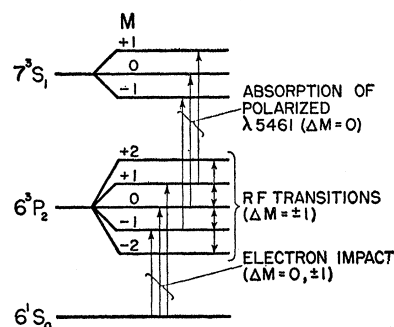

FIG. 1. Levels and transitions involved in paramagnetic resonance experiment on aligned metastable $^3P_2$ mercury atoms.

from the ground state. Details of a resonance experiment employing such double-hit ionization undertaken by I. Duck and the author will be reported elsewhere. A description of an experiment performed on Hg atoms in the metastable $^3P_2$ state will serve to illustrate the method and demonstrate the feasibility of the absorption scheme.

A planar diode containing Hg-vapor saturated at room temperature ($\sim 1 \times 10^{-3}$mm Hg) with a gap of 2 cm and a hot oxide cathode of 3 cm^2 area was operated at a current of about 200 ma and a plate voltage (<20 volts) favorable for the excitation of the $^3P_2$ state. Under these conditions, the gap is filled by an equipotential plasma and the cathode closely surrounded by an ion sheath. The electrons receive all their acceleration inside this ion sheath and enter the plasma in a beam normal to the planar cathode.[5] Parallel to the beam, a magnetic field H_0 of about 8.3 gauss was applied, which splits the $^3P_2$ level into five sublevels about 17.2 Mc/sec apart in the absence of nuclear moments (Fig. 1). Of these, the $M=0$ and, because of electron exchange, also the $M=\pm 1$ levels are predominantly excited by the beam. The alignment is now monitored by the absorption of $\lambda 5461$ radiation from an external source which was focused through the gap of the excitation tube and suitable filters on a photocell. The absorption by the metastables of the radiation which corresponds to the $6\,^3P_2 \rightarrow 7\,^3S_1$ transition amounted to about 10%. When the radiation is polarized parallel to H_0, the selection rule $\Delta M = 0$ applies and the absorption is mainly due to transitions from the $M=0$ level and to a lesser degree from the $M=\pm 1$ levels. This situation is appreciably changed when a rf field H_0 of a few milligauss or more and the proper frequency is applied perpendicular to H_0. $\Delta M = \pm 1$ transitions are induced between the magnetic sublevels, and the non-absorbing $M=\pm 2$ levels are populated at the expense of the $M=0,\pm 1$ levels which weakens the absorption. With the modulation techniques commonly used in paramagnetic resonance experiments, the variation of the absorption with the field H_0 can be displayed on an oscilloscope (Fig. 2). The g factor of the resonance was found by comparison with the signal from a K-NH$_3$ solution[6] to be 1.48 ± 0.01, in agreement with the optical value.
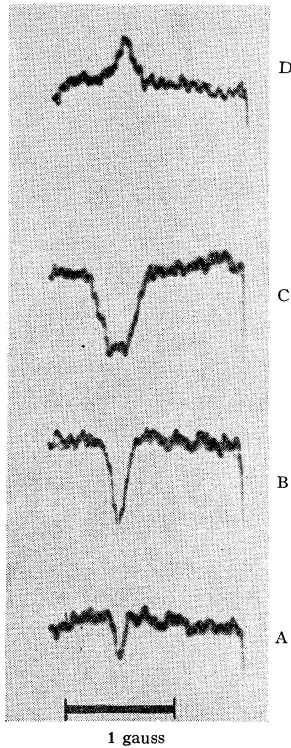
FIG. 2. Oscilloscope traces of λ5461 absorption by $^3P_2$ mercury atoms *versus* field H_0. A shows the decrease in absorption by paramagnetic resonance reorientation induced by a rf field of ∼12.5 milligauss, B and C are for ∼62.5 and ∼250 milligauss. Radiation polarized parallel to H_0 was employed for A−C. D shows the increase in absorption when polarization perpendicular to H_0 was used; rf field ∼62.5 milligauss.

1 gauss

Two weaker resonances corresponding to the $F=3/2$, $5/2$ states for the Hg^{199} isotope have also been observed at the expected field values. The extensions of the method to observe $\Delta F=\pm1$ transitions for the odd isotopes which would allow a precision determination of the hfs are obvious.

The author wishes to express his appreciation to his colleagues for advice and equipment, and to Mr. D. A. Schow and Mr. J. Jonson for the construction of the potassium resonance equipment and of various excitation tubes, respectively.

[1] H. W. B. Skinner, Proc. Roy. Soc. (London) A112, 642 (1926); H. W. B. Skinner and E. T. S. Appleyard, Proc. Roy. Soc. (London) A117, 224 (1928).
[2] J. R. Oppenheimer, Z. Physik 43, 27 (1927); H. Bethe, *Handbuch Der Physik* (Verlag Julius Springer, Berlin, 1933), Vol. 24, Part 1, p. 508.
[3] K. H. Kingdon, Phys. Rev. 21, 408 (1923).
[4] J. Brossel and F. Bitter, Phys. Rev. 86, 308 (1952).
[5] I. Langmuir, Phys. Rev. 33, 954 (1933).
[6] C. A. Hutchinson and P. C. Pastor, J. Chem. Phys. 21, 1959 (1953).

Ferroelectricity in the Langbeinite System*

F. JONA AND R. PEPINSKY

X-Ray and Crystal Analysis Laboratory, Department of Physics, The Pennsylvania State University, University Park, Pennsylvania

(Received June 29, 1956)

OPTICAL examination of $(NH_4)_2Cd_2(SO_4)_3$ over the range from room temperature to 77°K revealed a crystal transition about 10° above the lowest
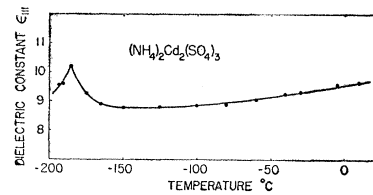

FIG. 1. Dielectric constant of $(NH_4)_2Cd_2(SO_4)_3$ as function of temperature, measured along cubic [111] direction.

temperature. The material is cubic above the transition point, and belongs to the Langbeinite $[K_2Mg_2(SO_4)_3]$ family. An x-ray powder pattern establishes the lattice constant as $a=10.35_0\pm0.005$ A. Growth from water solution at 80°C results in predominance of octahedral (111) faces.

Dielectric measurements were made on plates cut perpendicular to the cubic [111] direction. The behavior of the low-field dielectric constant ϵ_{111} as a function of temperature is depicted in Fig. 1. The room-temperature value of ϵ_{111} is about 9.5; the constant begins to climb slightly at −160°C, and reaches a peak of 10.2 at −184°C. Below this point, ferroelectric hysteresis loops are observed. The coercive field at −190°C is approximately 15 kv/cm for an applied field of 25 kv/cm, and the spontaneous polarization above the former [111] cube direction is about 0.3 microcoulomb/cm². The hysteresis loops sometimes appear with noncentric symmetry, as in the case of guanidinium aluminum sulfate hexahydrate as reported by Holden et al.[1]

Detailed dielectric, optical, thermal, and x-ray measurements on this and isomorphous crystals are in progress.

[1] Holden, Merz, Remeika, and Matthias, Phys. Rev. 101, 962 (1956).

Effects of Superexchange on the Nuclear Magnetic Resonance of MnF₂

R. G. SHULMAN AND V. JACCARINO

Bell Telephone Laboratories, Murray Hill, New Jersey

(Received June 29, 1956)

NUCLEAR magnetic resonances of F^{19} in oriented single crystals of MnF_2 have been observed at 77°K, 195°K, and 310°K. Two broad F^{19} resonances were found (corresponding to the two nonequivalent fluorine sites in the unit cell) whose separation shift from the F^{19} resonance in a diamagnetic material, were functions of the external magnetic field, crystal orientation, and temperature.

Bloembergen and Poulis[1] have pointed out that for crystal orientations such that H_0 is contained in the a-b plane, two F^{19} resonances are to be expected while only one is to be found for H_0 in the b-c plane. With the former orientation we have observed the separate lines at 77°K, while at 310°K the lines were not resolved.
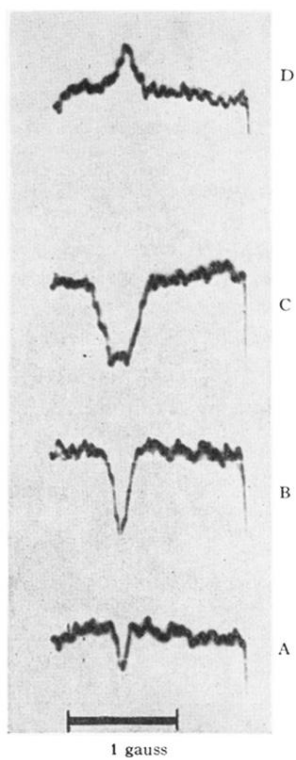
FIG. 2. Oscilloscope traces of λ5461 absorption by $^3P_2$ mercury atoms *versus* field H_0. *A* shows the decrease in absorption by paramagnetic resonance reorientation induced by a rf field of ∼12.5 milligauss, *B* and *C* are for ∼62.5 and ∼250 milligauss. Radiation polarized parallel to H_0 was employed for $A-C$. *D* shows the increase in absorption when polarization perpendicular to H_0 was used; rf field ∼62.5 milligauss.