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LIGHT MODULATION AT THE GROUND-STATE HYPERFINE-SEPARATION FREQUENCY OF POTASSIUM*

Arthur H. Firester and Thomas R. Carver
Palmer Physical Laboratory, Princeton University, Princeton, New Jersey
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We report the observation of light modulation at hyperfine frequencies in optically pumped potassium vapor. Light modulation effects of the Bell and Bloom,[1] and Dodd, Series, and Taylor[2] type have previously been limited to relatively low frequencies. Bloom,[3] and later Bouchiat,[4] had suggested the extension of these techniques to ground-state hyperfine frequencies. The original suggestion was made at a time when a lack of comprehensive light-modulation theory left it unclear what light polarization was required. This, together with the difficulty in the design of a photodetector capable of response at hyperfine frequencies, is probably the reason for no previous report of experimental success.[5]

If two or more atomic levels are coherent or phase related throughout a bulk sample, there appears an interference term in the electric dipole absorption or emission probability. This term oscillates with the same periodicity as the source of coherence and shows a resonant behavior at the frequency separation of the atomic levels. Coherence between atomic states has been established by means of radiofrequency fields,[1,2] pulsed light sources,[6] pulsed electron beams,[7,8] and light-polarization modulation.[9] In our investigations, the $F=2$, $m_F=1$ and the $F=1$, $m_F=1$ levels of the ground state of potassium were magnetically coupled by means of a radiofrequency field directed along the z axis, thereby establishing coherence.

A schematic of the apparatus is shown in Fig. 1. A set of mutually orthogonal Helmholtz coils cancels the earth's field and produces a net field of about 400 mG along the z axis. The source of potassium pumping light is an electrodeless rf discharge lamp. The D_2 line is removed by a commercial interference filter. After passing through a circular polarizer, the light is incident on an absorption cell containing potassium metal and a few centimeters of helium buffer gas. This cell is contained within an oven at 60°C. The oven also serves as a shielded enclosure for a loop tuned to 462 MHz and oriented to generate a magnetic field along the z axis. The transmitted light is focused upon a type-917 photocell, coupled to

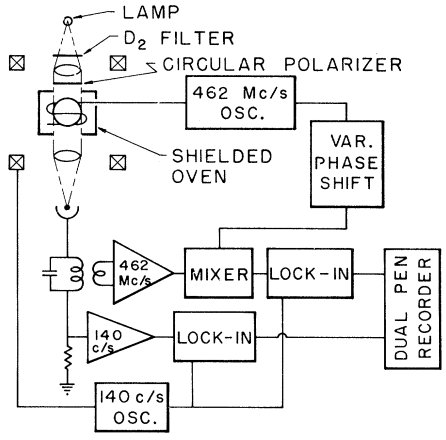

FIG. 1. A schematic diagram of the apparatus.

a tuned coaxial resonator having a $Q \approx 100$. This photocell was chosen because it is well suited to incorporation within a resonant structure. It has a wire anode and a cylindrical cathode. Connections to these two elements are made from opposite ends of the glass envelope. The photocell is operated at 500 V to reduce the effects of transit-time spread. The 462-MHz signal is amplified and mixed with a 462-MHz reference.[10] The reference oscillator also drives the loop about the absorption cell. The low-frequency output of the mixer is fed into a commercial lock-in detector, whose reference is derived from a modulation of the steady-state magnetic field at 140 Hz. Ten-second integration times were found to be desirable. In addition to the hyperfine detection system already described, a conventional detection system monitored the low-frequency variations of the transmitted light.

Both asymmetric and symmetric signals are predicted by a combination of time-dependent perturbation theory and a monitoring operator approach.[11] The signals have the general form of the two Bloch functions χ' and χ''. When viewed from a monitoring operator point of view, a necessary condition for the existence of light beats between two levels is that they are both connected to an excited state by the optical monitoring beam. The monitoring beam in our experiment is σ^+ circularly polarized light. The allowed optical transitions are illustrated in Fig. 2. Figure 3 is a tracing of the symmetric line. The lower trace is that of the zero-frequency detection system. The left-hand resonance is due to the $\Delta m = 0$ ($F = 2$, $m_F = 1$ to $F = 1$, $m_F = 1$) transition, while the right-hand resonance is due to a $\Delta m = 1$ ($F = 2$, $m_F = 2$ to $F = 1$, $m_F = 1$) transition. Had an rf field perpendicular to the z axis been used, the $\Delta m = 1$ resonance would have been many times larger than the $\Delta m = 0$ resonance. Careful design and orientation of the tuned rf loop eliminated most of the nonaxial fields. The $\Delta m = 1$ resonance attests to the presence of some remaining stray and fringing fields. It is evident (Fig. 2) that the $F = 2$, $m_F = 2$ and $F = 1$, $m_F = 1$ are not coupled to any common excited state, and thus there is no light-beat resonance (the upper trace in Fig. 3) corresponding to this transition. Both traces are derivatives of the actual line shapes as a consequence of the lock-in detection systems.

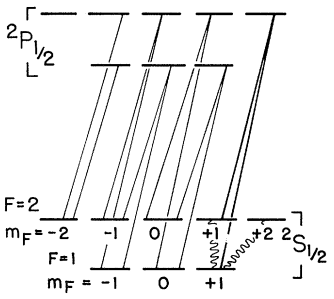

FIG. 2. A schematic of the ground and first excited states of potassium. All σ^+ optical transitions are indicated, with those required for the $\Delta m = 0$ resonance indicated by heavier lines.

These techniques could be applied to a measurement of the first-order field-independent $\Delta m = 0$, $m_F = 0$ to $m_F = 0$ transition. Earlier investigators[12,13] were obliged to make use of isotope filtering to (1) create a population difference between the two levels and (2) optically detect the transition. Light-beat techniques obviate the latter requirement since they do not require that the levels have unequal dipole absorption probabilities. The population difference could be produced by the use of a periodic rf pulse[14] to transfer some of the excess population of the $m_F = 2$ level to the $m_F = 1$ level. PIN photodiodes[15] and traveling-wave phototubes[16] are available which operate well into the gigahertz region, enabling the application of these techniques to higher frequencies, for example the ground-state hyperfine splittings of Rb^{87} and Cs^{133} at 6834 and 9192 MHz, respectively.

The existence of optical absorption implies the existence of optical dispersion. It is thus possible to extend these techniques to very high-frequency phase modulation of nonresonant light, as has been done in mercury at low frequencies.[17]

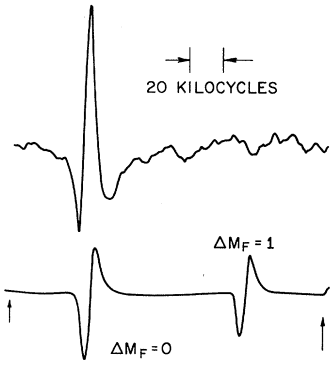

FIG. 3. The upper trace is the output of the 462-MHz light-beat detection system. An asymmetric line shape is shown. The lower trace is the output of the zero-frequency detection system.

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RELAXATION PHENOMENA IN MÖSSBAUER SPECTRA OF MAGNETICALLY ORDERED ERBIUM IONS IN $ErFeO_3$

I. Nowik and H. H. Wickman
Bell Telephone Laboratories, Murray Hill, New Jersey
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Recent low-temperature Er^{166} Mössbauer data, reported by Wiedemann and Zinn in a study of $ErFeO_3$, are interpreted in terms of spin-relaxation effects. This appears to be the first time relaxation effects have been observed in the Mössbauer spectra from a magnetically ordered system.

Wiedemann and Zinn[1] have recently reported unusual Mössbauer spectra for Er^{3+} in $ErFeO_3$, at sample temperatures below the Néel temperature (4.3°K)[2] of the erbium sublattice (the ferromagnetic iron-lattice Curie temperature is[3] 640°K). The Mössbauer transition employed was the $2^+ \rightarrow 0^+$, 80.6-keV gamma transition of Er^{166}. We show below that these spectra may be readily interpreted in terms of spin-relaxation effects. The $ErFeO_3$ system, therefore, appears to be the first reported example of such behavior in the Mössbauer-effect (ME) spectra from a magnetically ordered system.

The spin-relaxation interpretation differs from that of Wiedemann and Zinn, who suggested that there are several magnetically inequivalent Er^{3+} sites in $ErFeO_3$ and that the effective magnetic fields acting on the erbium nuclei in the various sites have different temperature dependences, thus leading to the observed spectra.[1] However, crystal structure,[4] magnetic structure,[2] and recent ME spectra[5] of Dy^{3+} in isomorphous $DyFeO_3$ argue for at most two magnetically inequivalent Er^{3+} sites. The magnetic and Mössbauer data are, in fact, well represented by a single magnetic rare-earth site. With this assumption relaxation spectra were computed and excellent agreement between experiment and theory was obtained.

In order to discuss the rare-earth hyperfine fields, it is first necessary to decide on a model for the pertinent electronic properties of the magnetic erbium ions. In the temperature range of interest, the erbium ions are exposed to an exchange field made up of two contributions: (1) an effectively constant exchange field from the iron atoms, and (2) at $T \leq T_C(Er^{3+})$, an induced molecular field in the erbium sublattice arising from exchange between the rare-earth ions. Near the transition temperature, the constant iron field dominates the erbium exchange field. The induced field in the erbium sublattice which gives rise to collective