Freezing out of hyperfine structure of the ground state of thallium atoms in a discharge

E. B. Aleksandrov and N. N. Yakobson

S. I. Vavilov State Optical Institute

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The "freezing-out" of the populations of the hyperfine structure of the ground state of Tl atoms under gas-discharge conditions was observed. The effective temperature of the ground state was 10 K at a vapor working temperature 1000 K.

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The atoms in a gas discharge are subject to intense resonant irradiation, which is the general case is not at equilibrium. One can therefore expect radiative exchange processes to cause deviations from thermal equilibrium in the distribution of the atoms over the sublevels of the long-lived states, and in particular over the sublevels of the hyperfine structure (hfs) of the ground state, if the distance between them is commensurate with the width of the resonance lines or larger. We have observed an appreciable deviation from equilibrium in the distribution of the populations of the hfs of thallium in a discharge, by using radio-optical and radio-spectroscopy methods.

A capacitive high-frequency (about 50 MHz) discharge in Tl vapor saturated at 600–650 °C, with a buffer gas (neon 3–10 Torr), was excited in a quartz thin-wall tube of inside diameter 4 mm. The tube was made coaxial with a cylindrical heat-resistant stainless steel microwave resonator tuned to 21 310 Hz in the TE_{011} mode. The resonator was excited through a coupling port in the sidewall, and had a loaded Q of 10^3. The discharge radiation was excited through a limiting aperture in the massive wall of the resonator. The source of the microwave power was a G 3-91 generator. The generator frequency was square-wave modulated with a deviation 50–100 kHz at a frequency 10^3 Hz and was slowly scanned in the vicinity of the frequency of the transition $\Delta F = \pm 1$, $\Delta m_F = 0$ of Tl^{205}. The frequency was calibrated, with accuracy 10^{-7}, with a quartz-

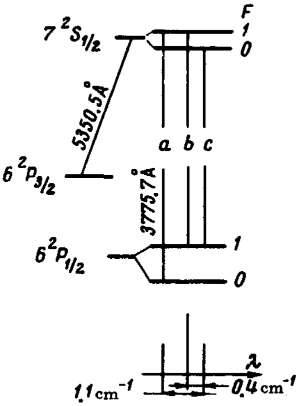

FIG. 1. Diagram of the three lowest terms of Tl (the theoretical intensities of the fine-structure components are indicated).

controlled heterodyne wave meter. In the radio-optical experiment, the registration was with the aid of a photomultiplier, using the hfs components of the 3776 Å line separated by a Fabry-Perot interferometer (Fig. 1), or else with the aid of a photocell integrally in the lines 3776 Å or 5350 Å separated with glass filters. The photosignal was amplified and synchronously detected in phase with the modulation of the microwave frequency.

In all cases, we registered a resonance signal in the hfs of the ground state. A typical plot of the signal in the case of integral registration in the 3776 Å line is shown in Fig. 2 (time constant in the registration channel 1 sec, width of signal significantly governed by the spectrum of the G 3-91 klystron). The three resonance components correspond to the structures of the transitions $F = 0$, $m_F = 0 \rightarrow F = 1$, $m_F = 0, \pm 1$ in the earth's magnetic field. The intensity ratio is determined by the orientation of the resonator axis relative to the magnetic-field vector. The central line exhibits saturation at a microwave power supply on the order of 10^{-4} W. In the case of the separate observation in the hfs components of the 3776 Å line, it was established that excitation of the transition $\Delta F = \pm 1$ leads to an increase of the intensity of component "a" and to a decrease of the intensity of the components "b" and "c" (Fig. 1). In the integral observation, the signal depends on the ratio of the intensity of hfs component, which changes with the thallium vapor pressure,[1,2] so that the 3776 Å resonance signal goes through zero and reverses sign at a certain temperature. The character of the variation of the signal in the components of the 3776 Å line offers evidence of an excess population of the lower $F=0$ of the hfs under the discharge conditions.

We were able to verify this conclusion independently by means of a direct radiospectroscopy experiment, by observing the absorption of the microwave power in the resonator in the vicinity of the atomic resonance (Fig. 3). When the discharge was turned off, no traces of a signal connected with the thermal population difference (0.1% at 1000 K) could be observed, despite the appreciable increase of the sensitivity. This fact together with the measurements of the absorbed power made it possible to estimate the excess population of the sublevel $F=0$ roughly at about 10%, corresponding to an effective "freezing out" of the atoms at 10 K.

As already mentioned, the microwave resonance was observed also in the 5350 Å green line. In contrast to the signals in the 3776 Å line, the intensity of the 5350 Å line at microwave resonance decreases in all cases, a fact for which there is no unequivocal explanation as yet. The maximum intensities of the signals in the 3776 and 5350 Å lines are close and reach a value 10^3 relative to the noise at integration times of 1 sec.

The impetus for our further research were studies[2] in which the anomaly, observed in[1], of the ratio of the intensities of the fine structure components of the 3776 Å line was interpreted as a

FIG. 2. Signals of the magnetic resonances $\Delta F = \pm 1$ in the 3776 Å line.

consequence and evidence of *inversion* of populations of the hfs sublevels of the state $6^2P_{1/2}$ in the discharge. In our investigations we have confirmed the existence of this anomaly (line "*a*" has a higher intensity than the line "*c*," and at temperatures 600–680 °C it exceeds also the line "*b*"—Fig. 1), and have established additionally that it is practically independent of the type of discharge, or of its power, or of the shape of the discharge vessel, or of the type and pressure of the buffer gas in the range 0–20 Torr. However, our remaining results reject the conclusions of[2], since it has turned out that the populations of the ground state are not inverted but are "frozen-out," and the anomaly of the intensities of the hfs components of the 3776 Å line is not the consequence of

FIG. 3. Signal of the resonance $m_F=0\leftrightarrow F=1, F=1, m_F=0$ in the microwave band.

the difference in the populations of the hyperfine sublevels. Indeed, saturation of the hyperfine transition does not eliminate the anomaly of the intensity distribution, as would be expected from the reasoning of[2], but to the contrary enhances this anomaly somewhat. This is also indicated by the fact that to obtain the population difference it is necessary to select the type and the pressure of the buffer gas: in our case neon at 6 Torr and helium at 10 Torr were optimal. The signals in argon were much weaker, and could not be observed at all in xenon. At the same time, as already noted, the anomaly of the intensities is practically independent of the type and the pressure of the buffer gas.

To our knowledge, no population redistributions sufficient to register microwave resonance has been observed under gas-charge conditions. The observed resonance uncovers a unique possibility of investigating the relaxation processes and the frequency shifts in the hfs of the ground state of the atoms under the discharge conditions. In addition, prospects arise of investigating the hfs of atoms not subjected to the standard optical-pumping procedure (for example, those whose resonance lines lie in the far ultraviolet). Finally, interest attaches to a detailed investigation of the mechanisms that lead to such substantial deviations of the system from thermal equilibrium.

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[2] Yu. I. Turkin, Opt. Spektrosk. **2**, 290 (1957); **7**, 10 (1959).