Inverse population in a paramagnetic crystal produced as a result of thermal excitation of the spin system by a pulsed magnetic field

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A maser effect at the spin sublevels of Co^{2+}, which is caused by direct conversion of the thermal energy of the crystal immersed in liquid helium in a coherent microwave radiation, has been observed for the first time in an experiment with a $La_2Mg_3(NO_3)_{12}\cdot 24H_2O$ (LaMN) crystal with Co^{2+} and Ce^{3+} impurities.

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As is known, the concept[1-4] of obtaining an inverse population by thermal excitation, which is closely connected with the concentration[3,5] of the quantum heat engine, assumes the creation of an artificial discontinuity between the temperatures of the individual subsystems of a complex quantum system as a result of an adiabatically rapid cooling or heating. Until now, these ideas have been realized only in gas-dynamic lasers (GDL). The extension of the principle of thermal excitation to condensed media and also to new frequency bands has obvious advantages.

This experiment, proposed in Refs. 6 and 7, can be roughly described as follows. A static magnetic field $\mathbf{H}=\mathbf{H}_0+\mathbf{H}_\perp$ is applied to a crystal that was cooled to a temperature $T_L \lesssim 2$ K; the field $\mathbf{H}_0$ is directed at a small angle θ_0 to the C_3 crystallographic axis, while $\mathbf{H}_\perp$ is perpendicular to C_3. This case corresponds to a thermally balanced distribution of the populations of the spin sublevels of ions, this is represented by the solid lines in Fig. 1. The values of H_0 and θ_0 were chosen in such a way that in the absence of $\mathbf{H}_\perp$ the Zeeman splitting $\delta_b(\mathbf{H}_0)$ of the main doublet of Ce^{3+}, which has a highly anisotropic g factor ($g_\parallel=0.0235$ and $g_\perp=1.8264$), is equal to $A/2$ [A is the hyperfine structure (HFS) constant of $^{59}Co^{2+}$ in the field $\mathbf{H}_0$], and the electron Zeeman splitting $\delta_a\equiv\delta_a(\mathbf{H}_0)$ of the cobalt ions falls within the range

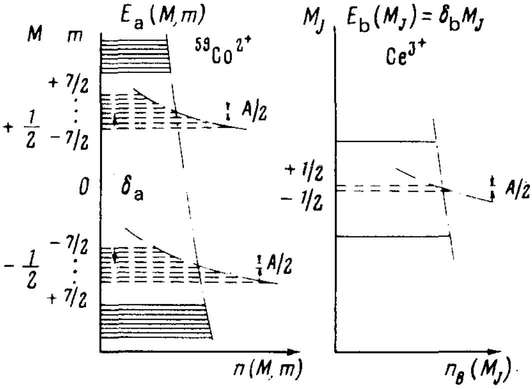

FIG. 1. Energy-level diagram and the population distribution of Co^{2+} and Ce^{3+} ions in LaMN in an external magnetic field.

$\lambda \approx 3$ cm. If the field $\mathbf{H}_\perp$ is now shut off in an adiabatically fast manner,[8] the spin temperature of the Ce^{3+} ion is reduced (to the value T_J) and a thermal contact is established between the sublevels of the cooled Ce^{3+} spins and the nuclear sublevels of $^{59}Co^{2+}$, which is caused by resonance cross relaxation at frequencies close to $A/2h$ (h is Planck's constant). If the cross-relaxation rates are significantly higher than the spin-lattice relaxation rates of Ce^{3+} and Co^{2+} and the density n_b of Ce^{3+} ions is much higher than the density n_a of Co^{2+} ions, then a short-term nonequilibrium distribution of populations, which is similar to that in Eq. (1), appears in the cobalt-sublevel system, because of thermal mixing[6,7]:

$$n(M,m) = \frac{n_a \exp(-\delta_a M/kT_S - AMm/kT_I)\sinh(A/4kT_I)}{2\cosh(\delta_a/2kT_S)\sinh[A(2\times I+1)/4kT_I]} \tag{1}$$

Here $n(M, m)$ is the density of atoms with a populated level (M, m) with an energy $E_a(M, m) = \delta_a M + AMm$, $I = 7/2$ is the nuclear spin of ^{59}Co, $M = \pm 1/2$, $-I \leqslant m \leqslant I$, and k is the Boltzmann constant. The quantities T_S and T_I in Eq. (1) are related by the relation $T_S \approx T_L > T_I \approx T_J$. This distribution is illustrated in Fig. 1 by the dashed lines. If the discontinuity between the temperatures T_L and T_J is so large that $AI/T_I > \delta_a/T_S$, then, according to Eq. (1), a partial inversion can occur and a maser effect is possible at least in the $1/2, -I \longleftrightarrow -1/2, -I$ transition.

The experiment was performed at a temperature $T_L = 1.5$ K with an LaMN sample grown from a solution with Co^{2+} and Ce^{3+} ion concentrations of 0.1 and 3 at.%, respectively. The Ce^{3+} spins were cooled by a pulsed field $\mathbf{H}_\perp(t)$, which was produced by the controlled discharge of a bank of condensers through a solenoid placed in the nitrogen flask of a helium cryostat. The field pulses had an approximately semi-sinusoidal shape (with a duration of 35 msec at the base) and an amplitude of 9 kOe. The electron paramagnetic resonance (EPR) spectrum of the Co^{2+} ions (X centers) was observed at a frequency of 9.127 GHz, and the sweep of the field H_0 was turned on at the end of the $\mathbf{H}_\perp(t)$ pulse. The initial H_0 value was slightly higher than the resonance field for the $1/2, -7/2 \longleftrightarrow -1/2, -7/2$ transition. It can be seen in the oscillogram of the EPR spectrum of Co^{2+} ions, which is illustrated in Fig. 2b,

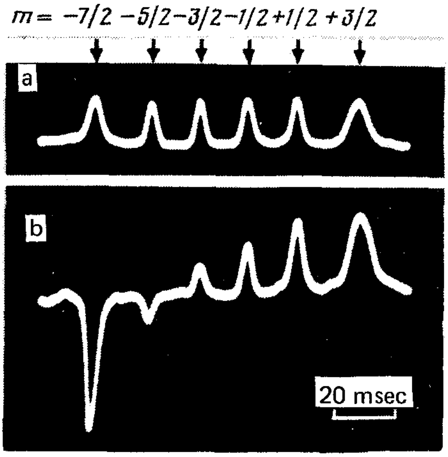

FIG. 2. Oscillograms of the EPR spectrum of Co^{2+} ions (a) at thermal equilibrium and (b) under conditions of thermal excitation of the spin system.

that under thermal-excitation conditions the two extreme HFS components (the $1/2, m \longleftrightarrow 1/2, m$ transitions with $m = -7/2$ and $-5/2$) are inverted. The inversion coefficients, determined from Fig. 2, are 3.1 and 0.6, respectively. The observed spectrum exhibited a strong angular dependence; the maser effect appeared only in the angular interval $|\theta_0| < 2.5°$. The time dependences of the induced radiation were also examined. The measurements were carried out at constant, resonance values of the field H_0; the observed curves were in qualitative agreement with those predicted theoretically.[6] The duration of the inversion at the $1/2, -7/2 \longleftrightarrow -1/2, -7/2$ transition was ~30 msec. There are reasons to assume that the maser characteristics of the material in question can be improved considerably by choosing the optimum concentrations of the Co^{2+} and Ce^{3+} ions[1].

Within the framework of the quantum heat engine there is a basic analogy[6,7] between thermal GDL and the system studied by us, the latter being closer to a partial-inversion GDL[4] in terms of its basic characteristics (thermal excitation and partial inversion). It is important to note that in our system, in contrast to the GDL, the essential discontinuity between the temperatures of the heat source (crystal lattice in liquid helium) and heat sink (Ce^{3+} spin system) is achieved with almost no change in the temperature of the heat sink, which has the maximum heat capicity.

In conclusion, we note that the method described above compares favorably with the methods[9] of obtaining inversion in an electron-spin system, which were discussed but not realized and which are based on a sudden reversal[10] of the external magnetic field. The latter methods do not utilize microwave pumping, but require the solution of a difficult problem involving a very rapid crossing of the intersection region of spin sublevels.[9]

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[1] The inversion effect can be enhanced by using a field $H_{\perp}(t)$ that is asymmetric in time, i.e., a field with a slow (~150 msec) rise and rapid (~30/2 msec) fall.

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