

ordered state has not been finally established. This cannot be the usual ferromagnetic state, since the relative spontaneous moment does not exceed 0.5 (Fig. 3). It is most probable that the arrangement of the spins corresponds to a ferromagnetic helix with variable pitch.

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### SELECTIVE LASER EXCITATION OF HIGH VIBRATIONAL LEVELS OF THE HCl MOLECULE

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1. The most important problem in the problem of applying laser radiation selectively to a substance is direct selective excitation of the molecule vibrational levels. So far, only the first vibrational level of the molecule has been excited with laser radiation, and the higher levels were populated by collisions between the excited molecules. Unfortunately, molecule collisions lead to a loss of the selectivity of the excitation. We report here the first successful experiment on direct selective excitation of the third vibrational level of the HCl molecule ( $E_{exc} = 1.04$  eV) by absorption of laser radiation at the second vibrational overtone of the molecule. The exceedingly high selectivity of such a process has made it possible to excite molecules having a definite isotopic composition.

2. The experiment consisted of selective optical pumping of the HCl molecule in the vibrational band  $v = 0 \rightarrow v = 3$ , using a high-power laser that was tunable in the IR region and detecting the excited molecules by means of IR luminescence of the vibrational transitions  $v = 3 \rightarrow v = 2$  and  $v = 2 \rightarrow v = 1$ . The experimental setup is shown in Fig. 1. To excite the HCl molecule to the  $v = 3$  level it is necessary to have radiation in the  $1.18 \mu$  region with a frequency adjustable in a range  $100 - 200 \text{ cm}^{-1}$ . To this end, a special laser set-up was constructed using a driving laser with a tunable amplifier and stimulated Raman scattering (SRS) frequency converter. The neodymium laser with

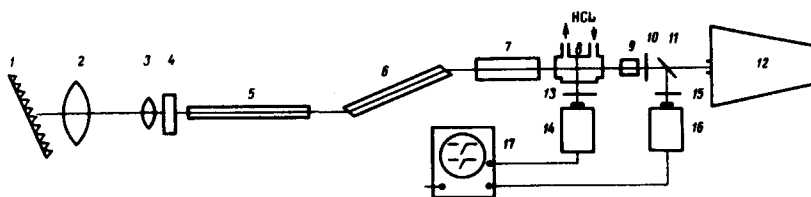


Fig. 1. Experimental setup: 1) diffraction grating, 2 - 3) telescope, 4) passive Q-switch, 5, 6) Nd rods, 7) cell with C<sub>6</sub>H<sub>5</sub>N, 8) cell with HCl, 9) KDP crystal, 10, 13, 15) filters, 11) beam-splitting plate, 12) STE-1 spectrograph, 14) Ge:Au receiver, 16) photomultiplier, 17) oscilloscope.

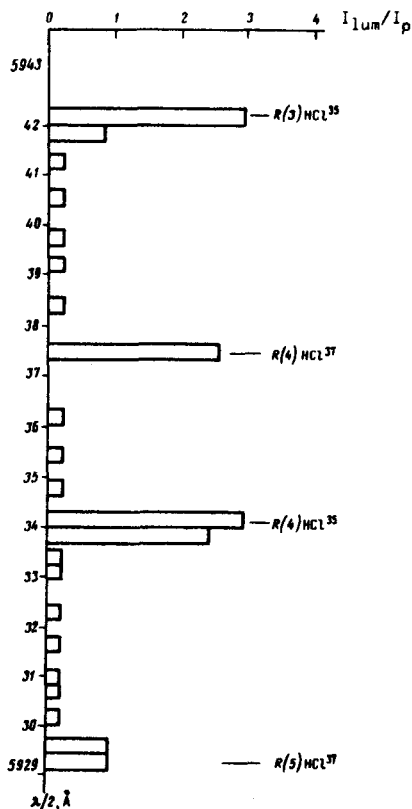


Fig. 2. Dependence of the luminescence-to-pump intensity ratio on the wavelength.

dye-solution Q-switch emitted a pulse of energy 1.0 - 1.5 J, duration 30 nsec, spectral width not larger than  $0.1 \text{ cm}^{-1}$ , and a frequency tunable within the width limits of the  $9410 \pm 100 \text{ cm}^{-1}$  neodymium gain line by means of a dispersion resonator. The resonator contained a diffraction grating (300 lines/mm) operating in third order. To increase the resolution of the lattice and to prevent its destruction, we used a telescope to increase the beam diameter from 10 to 60 mm. The laser output mirror was the end face of the active rod. Another rod outside the resonator was used to amplify the pulse energy to 2.5 - 4.0 J. The SRS in the cell with pyridine ( $\text{C}_6\text{H}_5\text{N}$ ) made it possible to mix the radiation frequency at  $990 \pm 2 \text{ cm}^{-1}$  in the region of the R-branch of the second overtone of HCl. The radiation at the first Stokes frequency had a duration 30 nsec, power 4 - 10 MW, and a spectrum width  $0.2 - 1 \text{ cm}^{-1}$ . The central frequency of this radiation was tuned by varying the frequency of the driving laser in the range  $8406.3 - 8434.1 \text{ cm}^{-1}$ , which spans the lines  $\text{R}(3)\text{HCl}^{37}$  ( $8407.3 \text{ cm}^{-1}$ ),  $\text{R}(3)\text{HCl}^{35}$  ( $8412.1 \text{ cm}^{-1}$ ),  $\text{R}(4)\text{HCl}^{37}$  ( $8418.0 \text{ cm}^{-1}$ ),  $\text{R}(4)\text{HCl}^{35}$  ( $8423.8 \text{ cm}^{-1}$ ), and  $\text{R}(5)\text{HCl}^{37}$  ( $8427.4 \text{ cm}^{-1}$ ) [1]. The wavelength of the IR Stokes component was registered with the large-dispersion STE-1 spectrograph, using the second harmonic produced by a KDP crystal.

The pump radiation passed through a quartz cell with HCl at a pressure 20 - 60 Torr. The cell had a transverse NaCl window transparent to the luminescence of the molecules in the vibrational transitions  $v = 3 \rightarrow v = 2$  and  $v = 2 \rightarrow v = 1$  in the region  $\lambda_{\text{lum}} = 3.5 - 4.0 \mu$ . A ger-

manium plate 1.5 mm thick was used to cut off the scattered radiation from the Stokes component  $\lambda_{\text{exc}} = 1.18 \mu$  and from the generator at  $\lambda_{\text{gen}} = 1.06 \mu$ . The luminescence was registered with a Ge:Au receiver with an approximate time constant of 2  $\mu\text{sec}$ . The intensities of the luminescence and excitation pulses were registered simultaneously with oscilloscopes.

3. We measured the dependence of the luminescence intensity on the pump wavelength while the driving laser frequency was varied by rotating the diffraction grating. Figure 2 shows this dependence obtained in one of the experiments after a series of 25 excitation pulses at different frequencies and at an HCl pressure 60 Torr. In the region of the absorption frequencies of  $\text{HCl}^{35}$  and  $\text{HCl}^{37}$ , designated by the arrows in Fig. 2, we see a resonant increase of the luminescence intensity, indicating that the HCl molecules with definite isotopic compositions are selectively excited to the third vibrational level. Since the width of the SRS momentum spectrum exceeds the width of the HCl absorption line ( $0.03 - 0.05 \text{ cm}^{-1}$ ), and the spectrum is not smooth, it follows that the ratio of the luminescence intensity in excitation at the absorption line of the isotopic molecules is equal to the ratio of the isotopes  $\text{Cl}^{35}$  and  $\text{Cl}^{37}$  in the natural mixture (3:1) only when averaged over a large number of pulses. In a series having a limited number of pulses, the results for which are shown in Fig. 2, we see only a tendency towards this value.

The luminescence pulse duration at 20 Torr is  $\tau_{lum} = 16$   $\mu$ sec. This value agrees with the time of vibrational relaxation  $\tau_{vib} = 1.6$   $\mu$ sec-atm for the HCl molecules in the  $v = 1 \rightarrow 0$  transition [2], if it is assumed that the relaxation rate increases with increasing number of the vibrational level.

We note that in this experiment the low level of the luminescence signal was due to the small absorption coefficient for the second overtone. Its value, say for  $R(3)HCl^{35}$ , is  $1.84 \times 10^{-3}$   $cm^{-2}atm$  [3], i.e.,  $1.2 \times 10^{-3}$   $cm^{-1}$  at 20 Torr and at an absorption line width  $0.04$   $cm^{-1}$ .

4. The described method for selective excitation of the high vibrational levels of an HCl molecule having a definite isotopic composition makes it possible to realize selective chemical reactions of molecules having an excess  $\sim 1$  eV of vibrational energy with other atoms and molecules, and also selective photodissociation of the excited molecules [4], whose red photoabsorption boundary is shifted by  $8000 - 9000$   $cm^{-1}$ .

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#### NONCOLLINEAR SPIN CONFIGURATIONS IN THE IRON GARNETS $Y_{3-x}Ca_xFe_{5-x}Sn_xO_{12}$

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In an analysis of the values of the magnetic moments of iron garnets with substituted nonmagnetic ions, Geller and co-workers [1] concluded that angular spin configurations can exist in such systems. Nowik [2] examined the relations between the exchange integrals of the intra- and inter-sublattice interactions and showed that the minimum energy corresponds to a noncollinear arrangement of the spins of those ions which have only one magnetic ion in their immediate surrounding, or have no magnetic neighbors at all.

We report here experimental observation of noncollinear configurations of the magnetic moments of  $Fe^{3+}$  ions in the d-sublattice of the substituted iron garnets  $\{Y_{3-x}Ca_x\}[Fe_{2-x}Sn_x](Fe_3)O_{12}$ .

We investigated the Mossbauer spectra of the  $Sn^{119}$  nuclei in this system of garnets for different substitutions ( $x = 0.1, 0.3, 0.5, 0.7, \text{ and } 0.9$ ) and temperatures (from  $4.2$  to  $600^\circ K$ ). We plotted the temperature dependence of the effective magnetic fields  $H_{eff}^{Sn}$  (T) at the tin nuclei (Fig. 1) and determined the values of  $H_{eff}^{Sn}$  at  $0^\circ K$ .

It was shown earlier that the magnetic field at tin ions placed in the a-sublattice of iron garnets is produced by the nearest  $Fe^{3+}$  ions in the d-sublattice [3 - 5].

The number of iron ions in the d-sublattices of the garnets  $\{Y_{3-x}Ca_x\}[Fe_{2-x}Sn_x](Fe_3)O_{12}$  does not change with increasing substitution. However, the