

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  $8000 - 9000$   $cm^{-1}$ .

In conclusion, the authors thank V.P. Kalinin for help with the work.

- [1] D.H. Rank, B.S. Rao, and T.A. Wiggins, J. of Molec. Spectr. 17, 122 (1965).
- [2] Hao-Lin Chen and Bradley Moore, J. of Chem. Phys. 54, 4072 (1971).
- [3] W. Benedict, R. Herman, and G. Moors, J. of Chem. Phys. 26, 1671 (1957)
- [4] R. V. Ambartzumian and V.S. Letokhov, IEEE J. of Quantum Electronics, QE-7, 305 (1971).

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

I.S. Lyubutin and A.P. Dodokin

Crystallography Institute, USSR Academy of Sciences

Submitted 11 February 1972

ZhETF Pis. Red. 15, No. 6, 339 - 343 (20 March 1972)

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

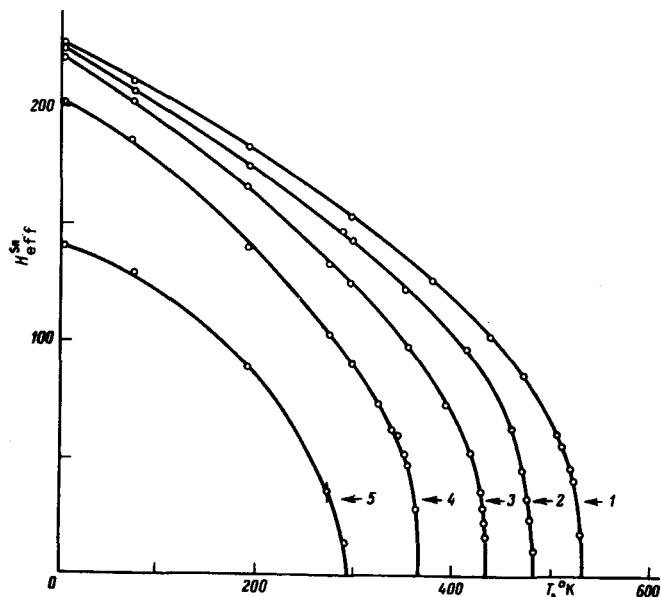


Fig. 1. Temperature dependence of the hyperfine magnetic fields at  $\text{Sn}^{119}$  nuclei in the iron garnets  $\{\text{Y}_{3-x}\text{Ca}_x\}[\text{Fe}_{2-x}\text{Sn}_x](\text{Fe}_3)_0_{12}$  for different  $x$ :  
 1)  $x = 0.1$ , 2)  $x = 0.3$ , 3)  $x = 0.5$ ,  
 4)  $x = 0.7$ , 5)  $x = 0.9$

iron d-ion occupy magnetically nonequivalent positions due to different nearest surroundings. There are altogether five nonequivalent d-positions, for which the numbers of the nearest magnetic  $\text{Fe}^{3+}$  ions in the a-sublattice are  $k = 4, 3, 2, 1$ , and 0. Only four types of nonequivalent iron d-ions lie around the tin ion ( $k = 4$  is excluded).

The contribution to the field  $H_{\text{eff}}^{\text{Sn}}$  at the tin from each type of nonequivalent iron d-ion is proportional to the average magnetic moment  $M_k$  of the given d-ion and to the probability  $P(k)$  of finding this type of d-ion in the nearest surrounding of the tin. The summary average field at the tin is the vector sum of all the contributions from four neighboring  $\text{Fe}^{3+}$  d-ions and coincides with the direction of the total magnetic moment of the d-sublattice [4]. The projections of the vectors  $M_k$  on this direction are specified by the coefficients  $\cos \theta_k$ , where  $\theta_k$  is the angle between the direction of the magnetic moment of the  $\text{Fe}^{3+}$  d-ion with  $k$  magnetic a-neighbors and the direction of the total d-sublattice magnetization.

The formula giving the value of the magnetic field at the tin for any concentration  $x$  and temperature  $T$  can be written in the form

$$H_{\text{eff}}^{\text{Sn}}(x, T) = H_{\text{eff}}^{\text{Sn}}(0, 0) \sum_{k=0}^3 a_k \cos \theta_k P(k) \bar{M}_k(T). \quad (1)$$

Here  $\sum_{k=0}^3 P(k) = 1$  and  $\bar{M}_k(T) = M_k(T)/M_k(0)$ . The coefficients  $a_k$  take into account the case when the mechanism whereby the magnetic field is induced at the tin ions by the iron ions is different for different nonequivalent  $\text{Fe}^{3+}$  d-ions [6].

Data on the crystal structure of the garnets show that the changes of the geometry of the nearest surroundings of the a- and d-ions as a result of the dimensions of the substituted ions are insignificant and amount only to fractions of one per cent [7]. In addition, experiments have shown that the values of the fields  $H_{\text{eff}}^{\text{Sn}}$  in yttrium and gadolinium iron garnets are the same [8], in

spite of some difference between the dimensions of the entering cations. Consequently, in our system of iron garnets we can neglect the change of the field  $H_{\text{eff}}^{\text{Sn}}$  due to distortions of the surroundings of the  $\text{Sn}^{4+}$  ions with increasing substitution. The coefficients  $a_k$  will in our case apparently be independent of the degree of substitution  $x$ , and should all be equal to unity:  $\alpha_3 = \alpha_2 = \alpha_1 = \alpha_0 = 1$ .

Formula (1) becomes particularly simple at  $T = 0^\circ\text{K}$ . In this case all  $\bar{M}_k = 1$  and the field at the tin, referred to zero concentration, is equal to

$$\bar{H}^{\text{Sn}} = \frac{H_{\text{eff}}^{\text{Sn}}(x, 0)}{H_{\text{eff}}^{\text{Sn}}(0, 0)} = P(3) \cos \theta_3 \quad (2)$$

$$+ P(2) \cos \theta_2 + P(1) \cos \theta_1 + P(0) \cos \theta_0.$$

Figure 2 shows an experimental plot of  $\bar{H}^{\text{Sn}}$  against  $x$  and the curves calculated from (2).

In the case of collinear arrangement of the spins of all the iron ions in the d-sublattice, the field  $\bar{H}^{\text{Sn}}$  should simply be assumed equal to the sum of all  $P(k)$  and should therefore be independent of  $x$  (dashed curve of Fig. 2). The experimental points, however, should deviate from such a plot. The change in the field  $H_{\text{eff}}^{\text{Sn}}$  as a result of the appearance of dipole contributions is excluded in this system of garnets. This is evidenced by measurements of the electric quadrupole interactions in magnetic fields at the iron nuclei as functions of the concentration [9]. Furthermore, at  $T = 0^\circ\text{K}$  all  $\bar{M}_k = 1$  and the surrounding of the Sn atoms remains magnetically symmetrical.

The observed experimental dependence of  $\bar{H}^{\text{Sn}}$  on  $x$  can be attributed only to violation of the collinearity of the magnetic moments of the  $\text{Fe}^{3+}$  ions in the d-sublattice.

The iron ions in the d-sublattice, which have no magnetic neighbors at all in the immediate surrounding, should have at low temperature a magnetic moment whose direction is nearly antiparallel to the total magnetization of the d-sublattice, owing to the antiferromagnetic intra-sublattice exchange interaction [10]. Their contribution to the field  $H_{\text{eff}}^{\text{Sn}}$  should therefore be negative.

The iron d-ions, which have one magnetic neighbor each in the a-sublattice, interact with one  $\text{Fe}^{3+}$  a-ion from the nearest surrounding and with four  $\text{Fe}^{3+}$  d-ions from the second coordination sphere. The relative orientation of the magnetic moment of such ions is determined by the competition of the energies of the inter-sublattice and intra-sublattice exchange interaction, the former tending to align the magnetic moments parallel inside the d-sublattice, and the latter antiparallel. As a result, the magnetic moment can make a certain angle with the direction of the d-sublattice magnetization [2].

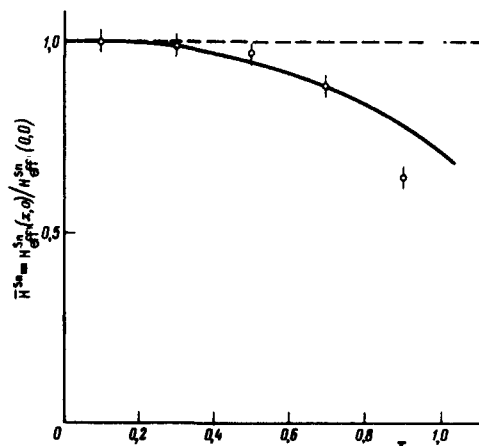


Fig. 2. Dependence of the effective magnetic field  $H_{\text{eff}}^{\text{Sn}}$  ( $0^\circ\text{K}$ ), referred to zero concentration, on  $x$ : points - experimental, curves - calculated with the aid of (2); dashed curve -  $\theta_3 = \theta_2 = \theta_1 = \theta_0 = 0^\circ$ , solid curve -  $\theta_3 = \theta_2 = 0^\circ$ ,  $\theta_1 = 63^\circ$ ,  $\theta_0 = 180^\circ$ ;  $\phi$  - experiment.

For the iron d-ions having two and more magnetic neighbors in the immediate surroundings, the inter-sublattice exchange interaction is dominant, and the magnetic moments of these ions are apparently collinear.

We have plotted several  $\bar{H}^{\text{Sn}}(x)$  curves calculated from (2) for different variants of the angles  $\theta_k$ . At definite values of  $\theta_k$ , the experimental  $\bar{H}^{\text{Sn}}(x)$  plot is satisfactorily described by formula (2) (see Fig. 2). Some deviations are due to the statistical character of the phenomenon in question and indicate that the angles  $\theta_k$  depend apparently somewhat on  $x$ .

For fixed  $x$ , from the experimental values of  $H_{\text{eff}}^{\text{Sn}}$  and with allowance for the assumptions made above we found the most probable values of the angles  $\theta_k$ :

$$\begin{array}{llll} x = 0.7 & \theta_3 = \theta_2 = 0^\circ, & \theta_1 = 63^\circ, & \theta_0 = 180^\circ. \\ x = 0.9 & \theta_3 = \theta_2 = 0^\circ, & \theta_1 = 108^\circ, & \theta_0 = 180^\circ \end{array}$$

Quite recently Bauminger et al. [11] analyzed the  $\gamma$ -resonance spectra of europium in europium iron garnets substituted with scandium, and concluded that noncollinear spin configurations probably exist in these garnets.

The authors are grateful to K.P. Belov and L.M. Belyaev for interest in the work, to M.P. Petrov for a discussion of dipole fields, and to A.Ya. Parshin for help with the experiments at helium temperatures.

- [1] S. Geller, H.J. Williams, G.P. Espinosa, and R.C. Sherwood, *Bell System Techn. J.* **43**, 565 (1964).
- [2] I. Nowik, *J. Appl. Phys.* **40**, 5184 (1969).
- [3] I.S. Lyubutin, V.A. Makarov, E.F. Makarov, and V.A. Povitskii, *ZhETF Pis. Red.* **7**, 370 (1968) [*JETP Lett.* **7**, 291 (1968)].
- [4] I.S. Lyubutin, *Proceedings of the Conference on the Application of Mossbauer Effect, Hungary, Tihany, 1969.*
- [5] I. Nowik, E.R. Bauminger, J. Hess, A. Mustachi, and S. Ofer, *Phys. Lett.* **34A**, 155 (1971).
- [6] I.B. Bersuker and I.Ya. Ogurtsov, *Zh. Teor. Eksp. Khim.* **4**, 48 (1968).
- [7] B.V. Mill', in: *Magnitnye i kristallokhimicheskie issledovaniya ferritov (Magnetic and Crystal-chemistry Investigations of Ferrites)*, Moscow State Univ. Press, 1971, p. 56.
- [8] I.S. Lyubutin, *Fiz. Tverd. Tela* **8**, 643 (1966) [*Sov. Phys.-Solid State* **8**, 519 (1966)].
- [9] I.S. Lyubutin, E.F. Makarov, and V.A. Povitskii, *Zh. Eksp. Teor. Fiz.* **53**, 65 (1967) [*Sov. Phys.-JETP* **26**, 44 (1968)].
- [10] I. Nowik, *Phys. Rev.* **171**, 550 (1968).
- [11] E.R. Bauminger, I. Nowik, and S. Ofer, *Phys. Lett.* **29A**, 328 (1969).

#### CAN LIQUID MOLECULAR HYDROGEN BE SUPERFLUID?

V.L. Ginzburg and A.A. Sobyenin  
 P.N. Lebedev Physics Institute, USSR Academy of Sciences  
 Submitted 24 January 1972  
*ZhETF Pis. Red.* **15**, No. 6, 343 - 345 (20 March 1972)

Any Bose liquid should apparently become superfluid at a certain temperature  $T_\lambda$ , provided it does not solidify at a higher temperature  $T_m > T_\lambda$ . This is corroborated by liquid helium as the example (it does not solidify at all at  $P < 25$  atm), and does not contradict the estimates for molecular hydrogen  $H_2$ . In fact, for an ideal Bose gas