[17]S. K. Kurtz, Acta Crystallogr. A 28, 5231 (1972).
[18]P. M. de Wolff, Acta Crystallogr. A 30, 777 (1974).
[19]A. Onodera and Y. Shiozaki, J. Phys. Soc. Jpn. 46, 157 (1979).
[20]T. S. Kuntsevich and N. V. Belov, Kristallografiya 16, 5 (1971) [Sov. Phys. Crystallogr. 16, 1 (1971)].
[21]V. F. Shabanov, A. N. Vtyurin, and S. Ya. Vetrov, Preprint IFSO-103F, Krasnoyarsk, 1979.
[22]F. F. Zernike and J. E. Midwinter, Applied Nonlinear Optics, Wiley, 1973, Chap. 2.

Translated by J. G. Adashko

Crystal growth control by electromagnetic radiation

G. M. Guro, G. A. Kalyuzhnaya, T. S. Mamedov, and L. A. Shelepin

P. N. Lebedev Physics Institute, USSR Academy of Sciences
(Submitted 14 May 1979)
Zh. Eksp. Teor. Fiz. 77, 2366–2374 (December 1979)

Gas epitaxy of narrow-band semiconductors irradiated by wide-spectrum lamps is experimentally investigated. It is shown that the application of definite doses of ultraviolet radiation accelerates the growth, improves the crystal quality, and lowers the epitaxy temperature. The mechanisms of the action of the radiation are investigated. Questions involved in the control of deviation from stoichiometry are considered. The possibility of using ultraviolet lasers for epitaxy is discussed.

PACS numbers: 81.10.Bk

1. The possibilities of nonthermal selective action of electromagnetic radiation on matter are receiving ever increasing attention of late. Much progress has been made in such applications of laser radiation as the stimulation of chemical reactions[1,2] and isotope separation.[3–5] The present paper is devoted to a new trend that has set in recently, namely the stimulating action of radiation on crystal growth.[6–10] This group of questions is of great importance in microelectronics, progress in which is closely connected with introduction of epitaxial technology methods in manufacture.

Following the first reports[6–9] of the use of broad-spectrum electromagnetic radiation for the growth of epitaxial semiconductor layers, many interesting results were obtained, particularly accelerated growth of epitaxial layers of germanium, silicon, and gallium arsenide[6–8] and of the tellurides of cadmium and lead.[9] However, attempts to use lasers were unsuccessful. Progress in this research was impeded to a considerable degree by the insufficient understanding of the mechanism of growth under irradiation conditions.

We report here experimental and theoretical research on the mechanisms of gas epitaxy of narrow-band semiconductors using xenon-lamp illumination, and determine the feasibility of the use of lasers. It is shown that under certain conditions ultraviolet (UV) irradiation increases the growth rates, improves the quality of the crystalline films, lowers the epitaxy temperatures, and affords a possibility of controlling the deviations from stoichiometry. A unique feature here is the strictly measured dose and the relatively low power of the UV irradiation. Thus, the use of lasers with properties that deviate from definite norms (with respect to intensity, spectral composition, and operating regime) is more or less useless.

2. Experiments on the epitaxial growth of layers 5 to 100 μm thick were performed on the compounds PbTe, $Pb_{1-x}Sn_xTe$, and HgTe under conditions of sublimation transport of the matter through the gas phase in a semi-enclosed volume of a quartz reactor in a hydrogen atmosphere. The light flux was used both to heat the reaction system (in conjunction with resistive heating) and for nonthermal action on the growth of the epitaxial layers. A diagram of the experimental setup is shown in Fig. 1. Among its distinguishing features is the presence of a radiation source, of an optical window in the reaction chamber, and of an optical system for the entry of the radiation into the reaction volume.

The radiation source was a DKSR-3000 ultrahigh-pressure xenon lamp emitting at 0.2 to 1.5 μm, with 10% of the power in the ultraviolet, 35% in the visible,

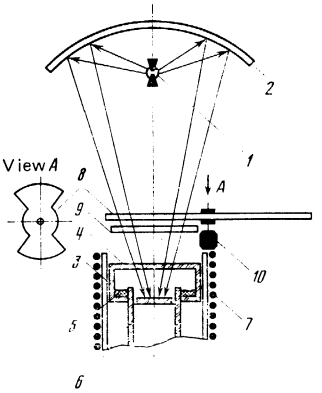

FIG. 1. Experimental setup for the study of the mechanism of the effect of optical irradiation on the growth of epitaxial semiconductor layers: 1—radiation source, 2—elliptic reflector, 3—quartz reactor, 4—optical-quartz window, 5—source of matter carried through the gas phase, 6—substrate, 7—resistance heater, 8—rotating shutter, 9—light filter, 10—motor.

 0038-5646/79/121141-05$02.40

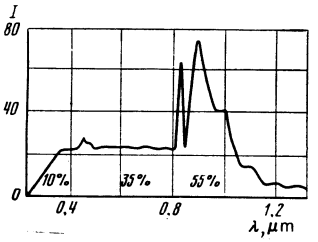

FIG. 2. Spectrum of radiation source, DKSR ultrahigh-pressure xenon lamp.

and 55% in the infrared. The spectrum of the radiation source is shown in Fig. 2. The quartz (grade KU-1 or KU-2) optical window in the reaction apparatus absorbed less than 1% of the radiation in the wavelength range 0.25–1.5 μm.

The spectral composition of the acting radiation was varied with light filters (Fig. 3). The substrates were single-crystal BaF_2 (111) plates finished in the same way in all the experiments. For any one deposited material, the substrate and source temperatures were maintained equal, regardless of the filter used, and were measured directly during the time of the experiment. The filters were placed outside the quartz reactor in such a way that the reacting gas medium and the substrate were subjected to the same radiant flux density in all experiments. The experimental results are given in Table I.

When no filters were used (wavelength band 0.25–1.5 μm) the growth rate v of the single-crystal perfect layers of PbTe and $Pb_{1-x}Sn_xTe$ was 2.2 μm/min, and that of HgTe was 2.5 μm/min. When UV in the 0.25–0.4 μm band was used (Fig. 3a) the epitaxial-layer growth rate increased sharply and reached 6 μm/min for PbTe and $Pb_{4-x}Sn_xTe$ and 15 μm/min for HgTe. However so fast a growth can not be controlled, i.e., the material is deposited not only on the substrate but also in the entire volume of the reaction chamber. When the irradiation wavelength is λ = 0.4–1.4 μm (Fig. 3b–d) the growth rates of the epitaxial layers of the mentioned compounds decreases to 0.5–0.7 μm/min. Without irradiation, the growth rate drops to 0.1–0.2 μm/min. It is seen from the presented data that light increases substantially the growth rate of the epitaxial layers.

A study of the epitaxial growth in UV radiation (Fig. 4) has shown that a single-crystal layer grows only in a narrow energy range. When the UV intensity is decreased the growing layers are polycrystalline. When the UV intensity is increased the growth rate also increases, but a polycrystal is produced, and with further increase of the intensity the growth goes out of control.

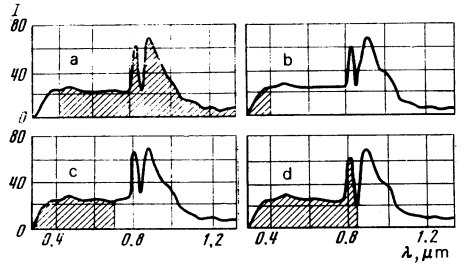

FIG. 3. Change of spectral composition of radiation source with the aid of light filters: a—transmission region 0.25–0.4 μm; b—0.4–1.4 μm; c—0.7–1.4 μm; d—0.82–1.4 μm.

TABLE I. Growth rate and crystal structure of grown PbTe, $Pb_{1-x}Sn_xTe$ and HgTe layers under optical illumination at intensity 100 W/cm².

Radiation band, μm	Material	v, μm/min	Layer structure
0.25–1.4	PbTe, $Pb_{1-x}Sn_xTe$	2.2	single crystal
	HgTe	2.5	single crystal
0.25–0.4	PbTe, $Pb_{1-x}Sn_xTe$	6	uncontrollable growth
	HgTe	15	uncontrollable growth
0.39–1.4	PbTe, $Pb_{1-x}Sn_xTe$	0.53	single crystal
	HgTe	0.83	single crystal
0.7–1.4	PbTe, $Pb_{1-x}Sn_xTe$	0.72	single crystal
	HgTe	0.5	single crystal
Irradiation of gas phase only with λ = 0.25–1.4 μm	PbTe, $Pb_{1-x}Sn_xTe$	0.35	polycrystal
Epitaxy without irradiation	PbTe, $Pb_{1-x}Sn_xTe$	0.2	mosaic single crystal
	HgTe	0.1	mosaic single crystal

In the experiments when only the gas was irradiated without filters and no radiation was incident on the substrate, the growth rate decreased sharply from 2.2 to 0.4 μm/min.

A study of the time dependence of the thickness d of the epitaxial film irradiated at λ = 0.25–1.4 μm on an insulating BaF_2 substrate has shown that in all cases there is an initial growth stage when the layer is not yet solid (Fig. 5) and then the effect of the radiation is small, and only after the end of this period does the growth rate increase considerably under auto-epitaxy conditions.

The noted experimental features of the epitaxial growth under the influence of radiation are apparently of general importance. Thus, electromagnetic radiation was used in epitaxy of silicon by the method of hydrogen reduction of silicon tetrachloride. Under the experimental conditions, the $SiCl_4 + H_2$ mixture absorbed 15–25% of the visible and UV radiation (the wavelength corresponding to the dissociation of $SiCl_2$ is λ = 0.316 μm). Kumagawa[8] has shown that the rate of silicon growth by hydrogen reduction from the tetrachloride under UV irradiation at 850–900 °C was 25–30% higher than ordinary conditions (1200 °C), and that at 1200 °C the growth rate is practically independent of the irradiation. At the same time Medvedev and Ivanikova's[6] study of the effect of radiation on the growth rate in the 0.2–1.5 μm band has shown that at 1200 °C the growth rate increases fivefold. Thus, in the case of silicon epitaxy, the radiation affects the course of the chemical reaction.

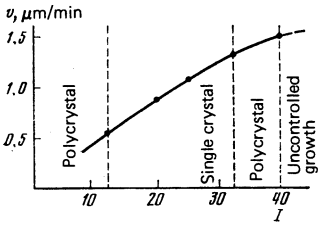

FIG. 4. Growth rate vs. UV radiation intensity.

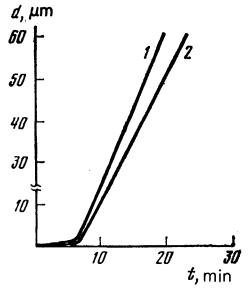

FIG. 5. Epitaxial-layer thickness vs. duration of process under irradiation at $\lambda = 0.25-1.4$ μm: curves 1 and 2 are for HgTe and PbTe, respectively.

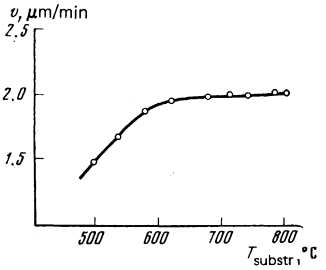

FIG. 6. Growth rate of epitaxial PbTe layers vs. substrate temperature when irradiated at $\lambda = 0.25-0.4$ μm.

3. In addition the accelerated crystal growth, and important experimental fact is the improvement of the quality of the epitaxial layers grown under irradiation conditions. It is seen from Table I that epitaxial growth of layers without irradiation, other technological conditions being the same, can result only in mosaic imperfect layers. The degree of perfection of the crystal structure of the epitaxial layers grown under irradiation conditions was estimated from the dislocation density. To this end, investigations were made of epitaxial $Pb_{0.78}Sn_{0.22}Te$ layers (including some doped with indium at various concentrations N_{In} during the time of growth under irradiation with light). The substrates were single-crystal plates of PbTe (100), $Pb_{0.79}Sn_{0.21}Te$ (100), BaF_2 (100), (111). Single-crystal layers up to 100 μm thick had the same orientation as the substrate. The dislocation density D in these layers was estimated from the density of the etch pits. The results are given in Table II.

As a result of the features of the growth under optical irradiation, the dislocation density in the films was much lower in all cases than in the PbTe or $Pb_{1-x}Sn_xT$ layers on BaF_2 (100) when exposed to light deconcentration of the indium introduced into the layer.

4. One of the important problems of modern epitaxial technology of semiconductors is the decrease of the temperature at which the layers are grown. Here, too, an important role can be played by the use of nonthermal action, primarily light. We have investigated, for the first time ever, the dependence of the threshold temperature of the epitaxy of narrow-band compounds of type A^4B^6 ($Pb_{1-x}Sn_xTe$) and A^2B^6 (HgTe) on the action of incoherent radiation in the 0.25–1.5 μm band. It was found that the growth temperature of single-crystal $Pb_{1-x}Sn_xTe$ and BaF_2 (100) when exposed to light decreases from 800 to 600 °C with practically no change in the rapid growth (see Fig. 6), and the threshold temperature at which the growth of the single crystal begins is 550 °C at a fixed irradiation intensity. Further increase of the temperature leads to growth of textured layers oriented relative to the substrate. Without irradiation and below 750 °C the growth is polycrystalline and its rate is lower by a decade than under irradiation conditions.

TABLE II. Dislocation density in epitaxial structures $Pb_{0.78}Sn_{0.22}Te$+In (layer)/$Pb_{0.79}Sn_{0.21}Te$ (substrate) and $Pb_{0.78}Sn_{0.22}Te$+In (layer)/PbTe (substrate) under condition of optical illumination at $\lambda = 0.25-1.4$ μm.

Epitaxial layer	Substrate	Substrate orientation	D_{substr}, cm^{-2}	D_{layer}, cm^{-2}	$D_{trans.layer}$, cm^{-2}	N_{In}, at.%
$Pb_{0.78}Sn_{0.22}Te$+In	$Pb_{0.79}Sn_{0.21}Te$	(100)	$5\cdot10^4$	$1\cdot10^4$	$5.2\cdot10^4$	0.4
$Pb_{0.78}Sn_{0.22}Te$+In	$Pb_{0.79}Sn_{0.21}Te$	(100)	$5\cdot10^4$	$2.5\cdot10^4$	$6\cdot10^4$	0.7
$Pb_{0.78}Sn_{0.22}Te$+In	PbTe	(100)	$1\cdot10^5$	$1\cdot10^4$	$1.5\cdot10^5$	0.2
$Pb_{0.78}Sn_{0.22}Te$+In	PbTe	(100)	$1\cdot10^5$	$8\cdot10^4$	$5\cdot10^5$	1.2

5. We proceed to examination of the mechanisms of epitaxial-layer growth. Under ordinary conditions without irradiation, the processes that occur can be described by the following qualitative picture. The growth surface of the semiconductor (PbTe, $Pb_{1-x}Sn_xTe$, HgTe, CdTe, Si) is impinged upon by atoms and molecules (the respective pairs Pb and Te_2; PbTe, SnTe; Hg and Te_2; Cd and Te_2; $SiCl_4$). The atom assumes its place in the lattice of the bonds of the supplied Te_2, PbTe, SnTe, $SiCl_4$ are broken The probability of such a process is relatively low (it increases with increasing substrate temperature). An analogy exists here with the gas-phase reactions that are characterized by the Arrhenius law (with a definite activation energy). A large number of collisions between the molecule and the surface is needed for the corresponding atom to land in its place in the lattice.

An estimate of the number z of the collisions can be obtained from the relation

$$z^{-1}\rho_g v_{mol} = \rho v,$$

where ρ and ρ_g are the numbers of crystal and gas molecules per unit volume, v_{mol} is the average velocity of the gas molecule, and v is the experimentally measured crystal growth rate. Under the experimental conditions $v = 0.2$ μm/min, $\rho = 10^{22}$ cm^{-3}, $v_{mol} = 1.6\times10^4$ cm/sec, and $\rho_g = 6.8\times10^{14}$ cm^{-3}. It follows therefore that only one out of 10^3–10^4 molecules colliding with the surface of the growing crystal will land in place. It is important that the process requires an activation energy.

In contrast to the reactions between saturated molecular compounds, reactions in the gas phase with participation of free atoms and radical are sub-barrier in character, and their cross sections are of the order of the gaskinetic ones.[11] In view of the sub-barrier character (similar to the gas-phase reactions) of the attachment of the free atoms to the surface, the crystal-growth conditions should improve noticeably with increasing number of free atoms.

Experiment has shown that the principal role in the acceleration of the crystal growth is played by the UV

part of the radiation. It is this part which is primarily responsible for the formation of free atoms in a gas of diatomic molecules ($E_{diss}=2.8$ and 2.7 eV for Te_2 and PbTe, respectively). A rough estimate of the concentration N_A of the free atoms produced upon absorption of the UV radiation (with allowance for recombination and diffusion), yields at an intensity $I_{UV}\sim 10$ W/cm^{-3} a density $N_A\sim 10^{13}$ cm^{-3}.[12] Hence, taking into account the value $v\sim 3\times 10^6$ cm/sec measured under irradiation conditions, we find that the effective number of collisions with the surface, needed for the free atom to land in place in the lattice is ~10–50, meaning that there is no barrier for the crystal growth in the presence of free atoms. Thus, if the free atoms amount to ~1% of the number of molecules, the growth due to the atoms becomes decisive. Control experiments were made to determine the composition of the gas phase by measuring the optical density of the vapor. It was found that under conditions of photostimulated epitaxy the amount of tellurium is increased at the expense of the PbTe molecules that are dissociated by the ultraviolet.

It is seen from the experimental results (Fig. 4) that at definite intensities the single-crystal growth becomes polycrystalline, while polycrystalline growth changes to formation of flakes (uncontrollable rapid crystal growth in the volume around randomly produced crystallization centers). Both the polycrystalline structure and the formation of the flakes are due to the appearance of clusters[13] near the crystal surface. The principal role is played in this process by the free atoms produced under the influence of the light. The cluster growth centers are charged particles (less than 1% of the number of free atoms) produced in the volume by the radiation and concentrated near the surface. Owing to the polarization forces,[14] the probability of atom capture by the ions is large ($\sigma\sim 10^{-15}$ cm^2) and the cluster has a high probability of remaining whole. The presence of free atoms with unsaturated bonds also greatly increases the strength of the clusters. Therefore even relatively small concentrations of free atoms can lead to formation and rapid growth of the clusters.

The lifetime τ of the cluster increases with the density of the free atoms and decreases with increasing temperature. According to the experimental data, the characteristic time of formation of the lattice (the minimum time needed to grow one molecular layer of thickness $a\sim 5\times 10^{-8}$ cm) is $\tau'=a/v'\sim 10^{-2}$ sec for single crystals and $\tau''=a/v''\sim 10^{-3}$ sec for a polycrystal, where v' and v'' are the maximum growth rates of the single crystal and the polycrystal. At $\tau\ll\tau''$ the clusters play no role. At $\tau\lesssim\tau''<\tau'$ the density of the free atoms produced by the irradiation is such that the growth rate of the produced clusters is much lower than the growth rate of the crystal. When captured by the surface, these clusters become nuclei for the formation of individual crystallites in the polycrystal. At $\tau\gtrsim\tau''$ the growth rates of the clusters and of the crystal become commensurate, the cluster growth becomes uncontrollable, and flakes are produced. This was precisely the situation produced in an interesting experiment,[10] in which IR lasers were produced to accelerate the epitaxy and too large a number of free atoms was produced.

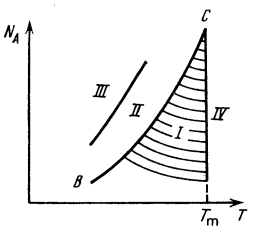

FIG. 7. Phase diagram on N_A-T plane. The curve BC corresponds to the condition of cluster formation. Region I—single-crystal state, II—polycrystal, III—chaotic phase (formation of flakes), IV—liquid phase. The curves in region I have constant epitaxy rates that increases with increasing T or N_A.

When the epitaxy temperature is lowered, the picture is analogous to that when the concentration of the free atoms is increased. According to our experimental data, single crystal growth takes place in the temperature interval from 780 to 500 °C and polycrystal growth below 500 °C. With further decrease of temperature (just as with increase of intensity) one can expect flakes to be produced. The observed decrease of the epitaxy rate (Fig. 6 is due to the decrease in the number of collisions with decreasing temperature. From the experimental data shown in Figs. 4 and 6 (just as from the condition for cluster formation[13]) follows the qualitative phase diagram shown in Fig. 7, which shows clearly the range in which the epitaxy regime is to be chosen.

We note that UV irradiation plays an essential role also in surface processes, by producing a surface charge on the growing semiconductor film.[15] Owing to the polarization forces that act on the molecules arriving at the surface, the strong electric field increases the growth rate in the breaks, corners, and in the troughs, thereby rectifying the surface defects.[12]

We have considered above free atoms produced under the influence of the radiation in the ground state. However, even a small admixture of excited atoms can play an important role in processes on a surface. The presence of this admixture can lead to a definite deviation from the stoichiometry of the growing crystal. Thus, the excited atoms can have valence properties that differ substantially from atoms in the ground state; if the excited states lies high enough, then the atom acts as a univalent one.

6. Our investigation points to concrete possibilities of technological use of lasers in microelectronics to act on the growth of epitaxial films. To increase the epitaxy rate and to improve the qualities of narrow-band semiconductors, as well as to grow silicon by decomposing the $SiCl_4+H_2$ mixture, it is necessary to use UV lasers with photon energies exceeding the dissociation energies of the supplied molecules ($\gtrsim 3$ eV). The laser intensities should be relatively low (~10 W/cm^2). When this intensity is exceeded, clusters and uncontrollable growth set in. We note that although laser radiation that raises the molecules to a region between dissociation and ionization can result in production of atoms and molecules in various states, one can speak of selectivity in the broad sense of the word.

In contrast to the pumping of vibrational levels of the molecules, we are dealing here with separation of definite aggregates of fast processes and acting on them during various stages of crystallization.

Besides gaseous media, great interest attaches to the action exerted on the degrees of freedom of a growing crystal by laser radiation at the frequencies of the atoms that make up the lattice. In this case the crystallization takes place in a state of an excited lattice that has properties (in particular stoichiometric) substantially different from those of the usual one. We note that in principle it is possible to use lasers to grow crystals in the liquid phase, by acting on definite chemical reactions that are connected with the crystallization process.[16]

[1]N. G. Basov, E. P. Markin, A. N. Oraevskiĭ, and A. V. Pankratov, Dokl. Akad. Nauk SSSR **198**, 1043 (1971) [Sov. Phys. Dokl. **16**, 445 (1971)].
[2]N. D. Artamonova, V. G. Platonenko, and R. V. Khokhlov, Zh. Eksp. Teor. Fiz. **58**, 2195 (1970) [Sov. Phys. JETP **31**, 1185 (1970)].
[3]N. V. Karlov and A. M. Prokhorov, Usp. Fiz. Nauk **118**, 583 (1976) [Sov. Phys. Usp. **19**, 285 (1976)].
[4]N. G. Basov, E. M. Belenov, V. A. Isakov, E. P. Markin, and V. I. Romanenko, Usp. Fiz. Nauk **121**, 427 (1977) [Sov. Phys. Usp. **20**, 209 (1977)].
[5]V. S. Letokhov and S. V. Mur, Kvantovaya elektron. (Moscow) **3**, 248, 485 (1976) [Sov. J. Quantum Electron. **6**, 129, 259 (1976)].
[6]S. A. Medvedev and G. E. Ivannikova, Elektronnaya tekhnika, ser. poluprovodnikovye pribory (Electronic Engineering, Semiconductor Devices) No. 4, 29 (1970).
[7]G. R. Gunter-Mohr, Patent USA No. 3.020.132.
[8]M. Kumagawa, J. Appl. Phys. Jpn. **7**, 1331 (1968).
[9]B. M. Vul, Yu. I. Gorina, G. E. Ivannikova, G. A. Kalyuzhnaya, S. N. Maksimovskiĭ, A. V. Kuznetsov, M. B. Nikiforov, V. I. Denis, V. I. Repshis, and M. I. Yarmalis, French Patent No. 2.335.043.
[10]A. D. Gerasimov, E. G. Zamkov, E. P. Markin, A. N. Oraevskiĭ, N. E. Ferapontov, and Yu. D. Chistyakov, in: Aktiviruemye protsessy v tekhnologii, (Activated Processes in Technology), No. 1, Taganrog, 1975, p. 59.
[11]V. N. Kondrat'ev and E. E. Nikitin, Kinetika i mekhanizm gazofaznykh reaktsii (Kinetics and Mechanisms of Gas Phase Reactions), Nauka, 1974.
[12]G. M. Guro, G. A. Kalyuzhnaya, T. S. Mamedov, and L. A. Shelepin, Preprint Fiz. Inst. Akad. Nauk SSSR No. 272, 1979.
[13]P. P. Kulik, G. E. Norman, and L. S. Polak, Khim. Vys. Energ. **10**, 203 (1976).
[14]B. M. Smirnov, Iony i vozbuzhdennye atomy v plazme (Ions and Excited Atoms in a Plasma), Atomizdat, 1974.
[15]V. G. Baru and F. F. Vol'kenshteĭn, Vliyanie oblucheniya na poverkhnostnye svoĭstva poluprovodnikov (Effect of Irradiation on the Surface Properties of Semiconductors), Nauka, 1978.
[16]N. V. Karlov, M. P. Stupin, and L. A. Shelepin, Pis'ma Zh. Tekh. Fiz. **2**, 828 (1976) [Sov. Tech. Phys. Lett. **2**, 323 (1976)].

Translated by J. G. Adashko

Interactions of nuclear spins in strongly anisotropic van Vleck paramagnets

A. V. Egorov, M. V. Eremin, M. S. Tagirov, and M. A. Teplov

Kazan' State University

(Submitted 25 May 1979)
Zh. Eksp. Teor. Fiz. **77**, 2375–2382 (December 1979)

Strong anisotropy of the second moment of the magnetic resonance line of ^{169}Tm nuclei in the dielectric van Vleck paramagnet $LiTmF_4$ (scheelite structure) has been theoretically observed and theoretically explained. The contributions made to the second moment of the absorption line by the interaction of the nuclear moments of the thulium with one another and with the nuclei of the diamagnetic ligands are calculated. It is shown that the dipole-dipole interaction of the nuclear magnetic moments of ^{169}Tm is effected via the $4f$-electron shells of the Tm^{3+} ions and is enhanced as a result by a factor $(1+\alpha_\perp)^2 \sim 5000$ times ($\alpha_\perp$ is the paramagnetic shift of the NMR line of thulium).

PACS numbers: 76.60. – k, 75.30.Gw, 75.20.Ck

During the last ten years, owing to the progress made in the techniques of obtaining infralow temperatures, a new trend has developed in solid state physics, involving research on nuclear magnetic ordering. Chapellier *et al.*[1] have performed brilliant experiments with the diamagnetic crystal CaF_2, in which they succeeded in attaining for the ^{19}F nuclei a spin temperature of the order of 10^{-7} K and in observing the antiferromagnetic ordering of the nuclear moments of fluorine. Andres, Bucher, *et al.*[4] investigated the series of intermetallic van Vleck paramagnets PrBi, $PrCu_2$, $PrCu_5$, in which the strong electron s–f exchange interaction shifted the temperatures of the magnetic ordering of the nuclear moments ^{141}Pr to the millidegree region. Finally, Bleaney and co-workers[5] were able to observe antiferromagnetic ordering of the nuclear spins of ^{165}Ho in the dielectric van Vleck paramagnet at a temperature 4.5 mK. In the planning of such experiments and in the development of methods for their realization one is inevitably faced with the question of preliminary esti-

 0038-5646/79/121145-05$02.40