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Quarterly Progress Report

Division 8

Solid State

15 January 1964

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Lincoln Laboratory

MASSACHUSETTS INSTITUTE OF TECHNOLOGY

Lexington, Massachusetts

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INTRODUCTION

This abbreviated report covers the work of Division 8 from 1 October 1963 through 31 December 1963. A more detailed presentation is covered by the Solid State Research report for the same period.

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Head, Division 8

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15 January 1964

This technical documentary report is approved for distribution.

Franklin C. Hudson, Deputy Chief
Air Force Lincoln Laboratory Office

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**1826	Magnetism and Crystal Structure in Nonmetals	J.B. Goodenough	Chapter 9, Magnetism: A Treatise on Modern Theory and Materials, H. Suhl and G.T. Rado, Editors (Academic Press, New York, 1963)
**2035	Cyclotron Resonance and Magneto-Optical Effects in Semiconductors	B. Lax	Proceedings of the International School of Physics Enrico Fermi (Academic Press, New York, 1963)
2041A	Propagation Constants for TE and TM Surface Waves on an Anisotropic Dielectric Cylinder	P.R. Longaker C.S. Roberts†	Trans. IEEE, PTGMTT MTT-11, 543 (1963)
2052	Criterion for Continuous Amplitude Oscillations of Optical Masers	J.I. Kaplan	J. Appl. Phys. 34, 3411 (1963)
2085A	Nernst and Seebeck Coefficients in Bismuth at High Magnetic Fields	T.C. Harman J.M. Honig	Advanced Energy Conversion 3, 525 (1963)
2089A	Galvano-Thermomagnetic Effects in Multi-Band Models	J.M. Honig T.C. Harman	Advanced Energy Conversion 3, 529 (1963)
2112	Directed Heating with Atoms, Electrons and Photons	T.B. Reed	ASM Technical Report No. NY9.1.62
2116	The Reaction of Rare Earth Oxides with a High Temperature Form of Rhodium (III) Oxide	A. Wold R.J. Arnott W. Croft†	Inorg. Chem. 2, 972 (1963)
2137	The Use of CO-CO_2 Atmospheres for the Preparation and Free Energy Determinations of Several Oxide Systems	W. Kunnmann D.B. Rogers A. Wold	J. Phys. Chem. Solids 24, 1535 (1963)
2138	Quantum Theory of Kinetic Equations for Electrons in Phonon Fields	P.N. Argyres	Phys. Rev. 132, 1527 (1963)

* Reprints available except where indicated by **.

† Author not at Lincoln Laboratory.

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2141	Second-Order Exchange Interactions from Spin Wave Resonance	R. Weber P. E. Tannenwald	J. Phys. Chem. Solids 24, 1357 (1963)
2143A	Optical Spectra of Several d^1-Electron Systems	J. R. O'Connor J. H. Chen*	J. Phys. Chem. Solids 24, 1382 (1963)
2153	Microwave Ultrasonics	P. E. Tannenwald	Microwave J. 6, No. 12, 61 (1963)
2155	Properties of GaAs Alloy Diodes	R. H. Rediker T. M. Quist	Solid-State Electronics 6, 657 (1963)
2197	Properties of InAs Diode Masers	I. Melngailis R. H. Rediker	Trans. IEEE, PTGED ED-10, 333 (1963)
2198	Index of Refraction of KDP	J. H. Dennis R. H. Kingston	Appl. Optics 2, 1334 (1963)
2220	High-Pressure Phases of Some Compounds of Groups II-VI	A. N. Mariano E. P. Warekois	Science 142, 672 (1963)
2232	Basic Science at the Crossroads	B. Lax	Microwave J. 7, No. 1, 16 (1964)
2243	High-Pressure Transitions in $A^{(III)}$ $B^{(VI)}$ Compounds. I. Indium Telluride	M. D. Banus R. E. Hanneman M. Strongin† K. Gooen*	Science 142, 662 (1963)
2246	Infrared InSb Laser Diode in High Magnetic Fields	R. J. Phelan A. R. Calawa R. H. Rediker R. J. Keyes B. Lax	Appl. Phys. Letters 3, 143 (1963)
2247	Semiconductor Lasers	B. Lax	Solid State Design 4, No. 11, 26 (1963)
2258	The Laser – A Long Range Future	B. Lax	Solid State Design 4, No. 11, 8 (1963)
2264	Two-Step Raman Scattering in Nitrobenzene	H. J. Zeiger P. E. Tannenwald S. Kern R. Herendeen	Phys. Rev. Letters 11, 419 (1963)
2280	Tunable Lasers	R. H. Rediker	Electronic Design 12, No. 1, 47 (1964)

* Author not at Lincoln Laboratory.

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2290	Magnetic Tuning of CW InSb Diode Laser	R.J. Phelan R.H. Rediker	Proc. IEEE (Correspondence) 52, 91 (1964)
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779B	Semiconductor Masers	B. Lax	Proceedings of the Symposium on Optical Masers (Polytechnic Institute of Brooklyn Press, 1963)
904	Recent Developments in Plasma Generation	T.B. Reed	Proc. National Electronics Conference, Northwestern University, 28-30 October 1963
910	Injection Lasers and Injection Luminescence	R.H. Rediker	NEREM Record, Boston, 4-6 November 1963
921	Nernst-Ettingshausen Energy Conversion	T.C. Harman	

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2193	Theory of the Faraday Effect in Solids	L.M. Roth	Accepted by Phys. Rev.
2202	High Pressure Transition in InSb	R.E. Hanneman M.D. Banus H.C. Gatos	Accepted by J. Phys. Chem. Solids
2219	Preparation and Properties of Sodium and Potassium Molybdenum Bronze Crystals	A. Wold W. Kunnmann R.J. Arnott A. Ferretti	Accepted by Inorg. Chem.
2223	Helium Temperature Ultraviolet Reflectometer for Use with Modified Spectrograph	W.J. Scouler	Accepted by Appl. Optics
2224	Current Regulator for Ultraviolet Light Source	W.J. Scouler E.D. Mills	Accepted by Rev. Sci. Instr.
2225	Reflectivity of HgSe and HgTe from 4 eV to 12 eV at 12° K and 300° K	W.J. Scouler G.B. Wright	Accepted by Phys. Rev.

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2241	A Ceramic Double Cell for Crystal Growth by Fused Salt Electrolysis	W. Kunnmann A. Ferretti	Accepted by Rev. Sci. Instr.
2253A	The Nernst-Ettingshausen Energy Conversion Figure of Merit for Bi and Bi-4%Sb Alloys	T.C. Harman J.M. Honig S. Fischler A.E. Paladino	Accepted by Solid-State Electronics
2254	Refined Treatment of the Theory Pertaining to Operating Characteristics of Anisotropic Nernst-Ettingshausen Devices	J.M. Honig B.M. Tarmy	Accepted by J. Appl. Phys.
2260	Bulk Solution of Ginzburg-Landau Equations for Type II Superconductor: Upper Critical Field Region	W.H. Kleiner L.M. Roth S.H. Autler	Accepted by Phys. Rev.
2265	Superconductivity in the High Pressure InSb-Beta Sn System	S.D. Nye M.D. Banus H.C. Gatos	Accepted by Appl. Phys. Letters
2271	Classical Ground State Spin Configurations in the Corundum Lattice	N. Menyuk K. Dwight	Accepted by J. Phys. Chem. Solids
2272	A Cluster Method for Finding Minimum Energy Spin States	D.H. Lyons* T.A. Kaplan	Accepted by J. Phys. Chem. Solids
2275	High Pressure Phase Transition in Tin Telluride	J.A. Kafalas A.N. Mariano	Accepted by Science
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2277	Theory of Spin Resonance and Relaxation	P.N. Argyres P.L. Kelley	Accepted by Phys. Rev.
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2293	Growing Helical Density Waves in Semiconductor Plasmas	C.E. Hurwitz A.L. McWhorter	Accepted by Phys. Rev.
2294	Recombination Radiation from Semiconductors	T.M. Quist	Accepted by Intl. Science and Technology
2302	Injection Luminescent Pumping of $CaF_2:U^{3+}$ Lasers	R.J. Keyes T.M. Quist	Accepted by Appl. Phys. Letters

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2303	2000-Ampere Pulse Generator	N. A. Sullivan	Accepted by Rev. Sci. Instr.
2306	Interferometric Phase Shift Techniques for Measuring Short Fluorescent Lifetimes	R. J. Carbone* P. R. Longaker	Accepted by Appl. Phys. Letters
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940	Narrow Band vs Localized d Electrons	J. B. Goodenough	Accepted Proc. Buhl Conf. on Materials, Pittsburgh, 31 October – 1 November 1963
941	Spin Correlations Among Narrow Band Electrons	J. B. Goodenough	Accepted by J. Appl. Phys. (Magnetism and Magnetic Materials Conference, Atlantic City, 12-15 November 1963)
943	Electrical Conductivity in the Spinel System $Co_{1-x}Li_xV_2O_4$	D. B. Rogers J. B. Goodenough A. Wold	

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924A	Semiconductor Injection Lasers	R. H. Rediker	New England Section, Optical Society of America, Waltham, 17 October 1963
934	Pressure Measurement and Efficiency in a Tetrahedral Anvil Press as Related to Anvil and Pyrophyllite Size	M. D. Banus S. D. Nye J. A. Kafalas	ASME Annual Meeting, Philadelphia, 17-22 November 1963
936	The $CaWO_4/Nd^{3+}(Na^+)$ Optical Maser: Wavelength and Polarization Dependence on Resonator Geometry	D. F. Edwards	American Physical Society, Chicago, 18-19 October 1963
938	Applications of Spark Source Mass Spectroscopy	E. B. Owens	Eastern Analytical Symposium, New York, 15 November 1963
957	Applications of Semiconductor Lasers	A. L. McWhorter B. Lax	First National Conference on Laser Technology, San Diego, 12-14 November 1963.
959	A Cluster Calculation of Magnetic Properties	G. F. Dresselhaus	Solid State Physics Seminar, Yale University, 18 October 1963
963A	Injection Lasers	R. H. Rediker	M. I. T., 25 October 1963

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965	InSb CW Diode Laser	R. J. Phelan R. H. Rediker	American Physical Society, Pasadena, 19-21 December 1963
983	Stoichiometry of Electronic Materials	A. J. Strauss	Industrial Liaison Symposium, M. I. T., 18 December 1963
987	Crystal Growth at Lincoln Laboratory	T. B. Reed	
984	Masers and Magneto-Optics	B. Lax	Optical Society of America, Chicago, 23-25 October 1963
1008	Injection Lasers	R. H. Rediker	Princeton University, 11 December 1963
1024	Stimulated Raman Emission	H. J. Zeiger	Physics Colloquium, Brandeis University, 19 January 1964

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I. SOLID STATE DEVICE RESEARCH

Forward-biased InSb diodes have been operated as CW lasers and the emission wavelength has been changed in steps of 50 Å from one cavity mode to an adjacent one by applying incremental steps of magnetic field of less than 600 gauss. The magnetic field also affects the refractive index n, as evidenced by the shift to shorter wavelength by 15 Å kgauss^{-1} of a given cavity mode. The measured 8-Å half-width of the emission line corresponds to the resolution of the grating spectrometer used. Continuous operation has been made possible at 2°K and in longitudinal magnetic fields above about 20 kgauss, by going to purer base material for the diodes. Threshold current densities for laser action at 2°K and 27 kgauss of 1400 amp cm^{-2} have been achieved.

A $CaF_2:U^{3+}$ laser has been pumped with the light input from GaAs injection lasers. Five GaAs injection lasers electrically connected in series were mounted along the slit of an integrating chamber in which was placed the 4 cm long by 3 mm diameter laser rod of $CaF:0.1\%U^{3+}$. Laser action at 2.631 microns was observed when the current through the diodes was increased above 3.7 amp, which corresponded to 2.9 watts of 0.84-micron radiation delivered to the integrating chamber. In injection luminescent pumping, the conversion efficiency of input electrical power into useful pump radiation may be as high as 50 percent. By using mixed semiconductor crystals, one can, in principle, tailor the luminescence wavelength to match a particular laser material ideally, and thus make the conversion of the pump radiation to laser radiation also very high. In addition to the higher efficiency per se, injection luminescent pumping would reduce the heat dissipation in the pumped laser by a factor of at least ten over that of flash-tube pumping. This reduction in heat dissipation should significantly reduce the thermal gradient problems that plague high-energy lasers. A high-efficiency system under study is $(Ga_xIn_{1-x})As$ diodes, radiating at 0.875 micron, pumping a Nd^{3+} laser which emits at 1.06 microns. Relatively large single crystals of $(Ga_xIn_{1-x})As$ have been grown by the halogen vapor transport mechanism in a closed system, and work is now in progress to approach the composition required to produce diodes which radiate at 0.875 micron.

The spectrum of the injection luminescence from InAs diodes at 300°K has been measured. The emission line occurs at 0.335 ev and has a half-width of 25 mev compared with the half-width of 12 mev at 2°K. The spectrum of the radiation from InAs diodes at 2°K has also been measured with a system that is more sensitive than that previously used. An additional peak is seen at 0.372 ev, 30 mev below the main emission peak, and a knee is observed at 0.34 ev.

A miniature "pill" laser package has been developed and consists of a GaAs laser and two semi-insulating GaAs spacers alloyed between two 0.080-inch-diameter metallic disks. Lasers in this package have been supplied to other groups in the Laboratory for use in optical radar development.

The flatness of diffused junctions in GaSb is being studied to determine whether irregularities in the junction plane can explain why lasers have not been fabricated from GaSb. It has been found for diffusions at 650°C from a dilute source of zinc in gallium that the junction plane more closely follows irregularities in the surface when the diffusant concentration is reduced.

Diffused p-n junction diodes have been fabricated of PbSe and PbTe. The procedure uses an interdiffusion mechanism for introducing controlled deviation from stoichiometry; excess lead gives rise to n-type and excess selenium and tellurium to p-type PbSe and PbTe, respectively. An analysis of the p-n junction depth results in the following effective interdiffusion constants: $D_{\Delta} = 4 \times 10^{-8}$ cm^2 sec^{-1} for producing a p-layer on n-type PbSe at 600°C; $D_{\Delta} = 9 \times 10^{-9}$ cm^2 sec^{-1} for an n-layer on p-type PbSe at 650°C; 6×10^{-7} cm^2 sec^{-1} for an n-layer on p-type PbTe at 650°C.

An experimental and theoretical investigation has been made of growing screw-shaped plasma density waves in a semiconductor bar subjected to parallel electric and magnetic fields. It is shown that in extrinsic material the growth is spatial, corresponding to stable traveling-wave amplification, whereas for nearly equal densities of positive and negative carriers, the wave is absolutely unstable and corresponds to the oscillistor phenomenon. Experimental observations were made in germanium and the growth rates and phase characteristics of the waves were found to be in excellent agreement with theory. For nearly intrinsic material, evidence of instability was found in accordance with the theoretical prediction.

A program has been started to investigate the current instability occurring in n-type GaAs at electric fields above 2000 volts cm^{-1}. The initial experiments of J. B. Gunn on the effects of light, temperature, and magnetic field on the instability have been extended to show that: light sufficient to change the sample conductivity does not affect the instability, immersing the sample in liquid nitrogen does not affeet the instability, and a 10,000-gauss magnetie field lowers the amplitude of the instability but does not change its threshold or spectrum.

II. LASER RESEARCH

The angular distribution data of stimulated Raman emission from nitrobenzene have been used to calculate the dispersion characteristics of nitrobenzene. Off-axis stimulated emission experiments are in progress to further substantiate the two-step Raman scattering model.

A theoretical study has been made of the growth of Raman radiation from noise. It is found that spherical-growing Stokes waves due to noise sources interact with the incident radiation to produee anti-Stokes radiation strongly peaked at the phase matching angle. The width of the anti-Stokes peak has been calculated.

New data on the optical absorption and electron spin resonance have been obtained for d^1 electrons in CaF_2. The results are explained by a strong, dynamical, Jahn-Teller distortion of the elcctron ground state.

Intense sources of visible pumping radiation are being developed for optically exciting GaAs. Repetitive pulsing at 13 pps and 5000 watts cm^{-2} should yield the required levels for study of possible laser action.

Short fluorescent lifetimes, such as those in GaAs, have been measured by an interferometric phase-shift technique. Samples are excited with multimode helium-neon laser radiation and the phase shift of the beat signal at 155 Mcps is used to determine the fluorescent decay time. Times in the 10^{-9}- to 10^{-10}-second region are easily measured, and typical values are reported for GaAs.

Multilayer dielectric reflectors have been produced separately and as coatings on GaAs lasers. As many as 17 layers have been deposited, and 99.9-percent reflection has been obtained over bandwidths as high as 1000 Å.

Improved ruby has yielded far-field diffraction patterns that approach perfect diffraction limited behavior.

III. MATERIALS RESEARCH

When a carrier gas is used in the vapor growth of crystals, usually the growth rate is inconveniently slow, since the carrier transports material from the source to the growing crystal by natural convection. The use of forced convection to increase the growth rate is being investigated by using this method to grow iodine crystals. Helium gas saturated with iodine vapor passes through a nozzle and strikes an air-cooled glass finger on which crystals are deposited. The rate and efficiency of crystal growth are being measured as a function of the temperature and helium flow rate.

A system has been constructed for making thermoelectric measurements at temperatures between −50° and 900°C on solids with resistances at least as high as 20 meg. Rigorous shielding of all parts of the system is necessary to prevent spurious readings due to pickup. The system has been used for measurements between 6° and 41°C on a crystal of Cr_2O_3-3%Al_2O_3 whose resistance increases from about 3 to 20 meg in this range. The Seebeck coefficient is $+900\,\mu v\ deg^{-1}$.

Thermoelectric measurements have been made on hot pressed polycrystalline CrO_2 at temperatures between −50° and 240°C. The values of Seebeck coefficient (-6 to $-24\,\mu v\ deg^{-1}$) and resistivity (1.2 to 4.3 milliohm-cm) are unusually low for an oxide. No clear evidence was found for a maximum in ρ with temperature, as previously reported in the literature.

Earlier electrical and optical measurements on n-type PbSe with carrier concentrations between 1×10^{19} and $2 \times 10^{20}\ cm^{-3}$ have been extended to n-type samples with electron concentrations as low as $1.5 \times 10^{18}\ cm^{-3}$ and p-type samples with hole concentrations between 3×10^{17} and $5 \times 10^{19}\ cm^{-3}$. Within the scatter of the data, the Hall mobilities at 4.2°, 77°, and 300°K and the thermoelectric power at 300°K are the same for n- and p-type samples with the same carrier concentrations. For both n- and p-type samples with sufficiently high carrier concentrations, the Hall coefficient increases between 77° and 300°K. The maximum value of R_{300}/R_{77} is 1.31 for p-type samples and 1.22 for n-type samples. The effective masses obtained by analysis of reflectivity data are somewhat higher for holes than for electrons, although the data for p-type samples are limited. The effective masses obtained by analysis of the thermoelectric power data on the assumption of acoustic lattice scattering and 4-ellipsoid models for both the

conduction and valence bands are somewhat smaller than the reflectivity values or published values based on Faraday rotation experiments.

The effect of NH_3 and H_2S adsorption on the spontaneous bending of thin {111} wafers of GaAs and InSb has been measured. For both these III-V compounds, adsorption of NH_3 decreases the curvature and elastic strain energy and adsorption of H_2S increases the curvature and elastic strain energy.

The system InSb-(β-Sn) has been investigated by measuring the superconducting transition temperature (T_c) as a function of composition for samples prepared by pressing mixtures of InSb and tin at 37 kbar and 450° to 480°C, quenching to room temperature or annealing at 300°C for one hour, and cooling to about −140°C before releasing the pressure. Treatment of the pure materials in this manner gives tetragonal $InSb_{II}$ (T_c = 2.1°K) or tetragonal β-Sn (T_c = 3.7°K). Alloying $InSb_{II}$ with only a few percent tin increases T_c to more than 4°K. A much smaller increase in T_c results from alloying InSb into β-Sn. The measurements indicate that the maximum solubility of β-Sn in $InSb_{II}$ is probably 2.5 to 5 atomic percent and that the maximum solubility of $InSb_{II}$ in β-Sn is probably about 35 atomic percent. In contrast, the maximum solubilities of β-Sn in normal InSb and of normal InSb in β-Sn are reported to be less than 0.5 and about 8 percent, respectively.

An analytical procedure is being developed for determining the three major constituents of HgS-HgSe alloys. After suitable sample treatment, mercury and selenium can be determined by the methods involving potentiometric titrations which were developed earlier for these elements. In initial experiments on the determination of sulfur in standard sulfate solutions, results accurate to within 1 to 2 percent have been obtained by an amperometric method based on titration with a lead solution.

IV. BAND STRUCTURE AND SPECTROSCOPY OF SOLIDS

Considerable progress continues to be made in understanding band structures of various solid state materials. Extensive calculations of the band edge structure of the compounds of lead with sulfur, selenium, and tellurium have been made. The results show that extrema of both the conduction and the valence bands probably lie at the (111) edge of the Brillouin zone and that there are six energy bands which influence these extrema. These six bands interact strongly; therefore, the properties of these compounds vary considerably, and the energy extrema are highly nonparabolic. The quantitative knowledge of the energy levels and Fermi surface of graphite is being improved as further experiments are made with more versatile experimental equipment and samples of higher purity and perfection. In pyrolytic graphite, a semimetal, several low-energy transitions have been seen with a behavior comparable to the "quantum effects" observed in the millimeter cyclotron resonance experiments in semiconductors at high magnetic fields.

The matrix elements in the quantum mechanical analysis of the Voigt effect have been modified to take into account second-order terms in the magnetic field. In germanium, 18 indirect interband transitions were observed in magnetoabsorption at 74 kgauss. Fine structure was also observed on some of these lines, and studies of this fine structure are now being carried out.

Experiments on the behavior of the excited levels of deep impurities with shifts of the band edges due to an applied uniaxial stress are in progress. It is expected that these experiments will clarify the nature of the infrared absorption lines which have been observed in silicon doped with sulfur.

There is also a strong continuing interest in both the theoretical and applied aspects of transport phenomena. The theoretical basis of transport is being broadened by work on generalized master equations, and transport theory under nonhomogenous conditions is being studied. From the standpoint of application, the properties of Nernst-Ettingshausen refrigerators are being analyzed with particular attention to the optimization of the refrigerator geometry. The properties of a device using single-crystal bismuth as a thermoelectric element have also been investigated.

V. MAGNETISM AND RESONANCE

Considerable progress has been made in flux techniques and electrolytic techniques for the growth of stoichiometric ferrites, chromites, and vanadites of the spinel structure and for the growth of a number of bronze crystals. A polytungstate flux has been versatile at permitting predictable results. Preliminary experiments on the systems MnAs-MnSb and MnAs-MnP appear to support our earlier suggestion that the first-order phase change at the Curie temperature of MnAs is due to a partial spin quenching that restabilizes the $B8_1$ phase.

Zero-field AFMR measurements as a function of temperature on powdered $YCrO_3$ give a Néel temperature of 129°K and a critical resonance frequency, extrapolated to 0°K, of ~198 kMcps. Measurements of the upper critical field of pure and impure niobium samples have been extended down to 0.4°K with the He^3 refrigerator. The temperature dependence of the attenuation of 70-kMcps longitudinal phonons in x-cut quartz has been measured from 4.2° to 25°K. A signal-to-noise ratio of 20 db has been obtained for the first echo pulse in the best quartz samples.

A theoretical investigation of the elastic magnetic scattering of neutrons at high temperatures has provided a model that permits interpretation of the anomalous "liquid" peaks sometimes observed experimentally (e.g., near the (111) nuclear position in $MnCr_2O_4$). In order to carry the analysis to lower temperatures, it will be necessary to work out higher terms of an expansion. An investigation of the first-order spin-orbit coupling of a d^5 configuration shows that this interaction gives a highly aspherical contribution to the spin density which increases the neutron diffraction intensity at the same peaks, and on the same order of magnitude, as the spherically symmetric Dzialoshinsky canting responsible for the weak ferromagnetic moment of α-Fe_2O_3. Hence this interaction is probably important for understanding the surprising, highly aspherical ferromagnetic spin-density distribution recently observed in α-Fe_2O_3 by Pickart, Nathans, and Halperin. The influence of magnetostriction on distortions from a spiral-spin configuration have been calculated for MnP. The analysis provides a richness of detail that, if verified experimentally, will permit a strong check for the spin correlations and energy-level diagram that have been assumed for this class of material. A hierarchy of approximations to the Heisenberg ferromagnet problem has been obtained. The method has been used to calculate the Curie temperature T_c, and the susceptibility and magnetization near T_c.