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Final Report

to the Army Research Office

ARO Proposal Number: P-16869-P Period of Contract: March 17, 1980 to September 15, 1983 Title of Contract: Spectroscopy of Thermal Energy Transport Contract Number: DAAG29-80-C-0085 Name of Institution: Indiana University Principle Investigator: W.E. Bron

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I. Research Program

During the lifetime of the contract the following research areas were undertaken:

A. Phonon Transport in the Presence of Quasidiffusion

A model experimental case was studied in which an originally monochromatic distribution of phonons are injected into a crystal and their transport in the presence of impurity scattering and anharmonic decay is determined. The experimental results did not agree with a simple analytical model of spatially segregated, temporally uncorrolated transport.

It turned out that we had observed for the first time a new mode of phonon transport, which has been termed "quasidiffusive" by two Russian theorists, Kazakovtsev and Levenson.

Quasidiffusive transport has the special properties that the time of flight response resembles pure diffusive motion but that the diffusion constant is not a constant of the motion but varies with the distance, L, between the source and the detector. Moreover, the phonon arrival time varies approximately linearly with L rather than with L^2 as is expected from pure diffusive flow.

This form of transport is not detectable by either thermal conductivity or by the heat pulse method. That is why it remained undetected until the more sophisticated transport experiments have become feasible. This transport mode should be quite general whenever impurity or isotopic scattering is strong, and should, for example, be a factor in thermal energy transport in some semiconducting devices. B. Generation of Coherent THz Phonons through Piezoelectric Surface Excitation

A long standing research project on piezoelectric surface excitation of coherent high frequency phonons has reached fruition during this period. It is shown that for such generation to be successful in the THz regime, stringent limitations must be observed in the degree of surface roughness and more importantly on subsurface damage. A key question is the existence of a subsurface damage layer which produces a finite gradient in the piezoelectric tensor over an extended region in the subsurface, leading thereby to interference effects between coherent phonons generated at different distances below the crystal surface.

The generation of hypersound in the THz region has been a long standing goal both in the basic physics of phonon generation and transport, but also in the applied area of ultrasonic testing, communication, etc.; the latter because of the higher band density available. It has been of considerable interest to U.S. Army research units. (See, for example, ECOM REPORT 4548 by J.R. Vig, J.W. LeBus, and R.L. Filler, and references cited therein.)

C. Phonon Lifetimes in the Picosecond Time Domain

We have developed a new application of a picosecond laser system to the measurement of the temperature dependence of the lifetime of LO phonons in GaP. The method permits a direct measurement of the phonon relaxation in the time domain. The results are compared to the traditional (indirect) determination

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of the lifetime from spontaneous Raman linewidths. A further comparison is made to the theoretically predicted temperature dependence of the lifetime. Limitations of the present picosecond system restrict similar comparisons for TO phonons to linewidth measurements. Good agreement is obtained among the experimental techniques and with theory.

The picosecond laser facility is used to produce packets of coherent LO phonons in GaP through stimulated Raman excitation. Natural decay of these phonons is observed by direct measurement of the intensity of a time delayable CARS signal. The nonlinear interaction of two dye laser pulse trains at frequencies ω_L and ω_S is used to resonantly drive a coherent set of Raman active phonons at $\omega_R = \omega_L - \omega_S$, and $\vec{k} \sim 0$. A low intensity component of the laser light at ω_L is temporally delayed through an optical delay line and is used to detect dephasing effects in the phonon packet.

The experimental observations lead the determination of T_2 ; the traditional <u>dephasing time</u>. The temperature dependence of T_2 is determined over a range from 5 to 300 K.

The time, T_2 , includes contributions from all dephasing processes such as elastic scattering from surfaces, from impurities, from imperfections, from photo and thermally excited electronic carriers, inhomogeneous line broadening, and also includes phonon-phonon scattering which leads to phonon population decay. (The population decay time is conventionally referred to as T_1). Good agreement between the theoretically obtained temperature dependence, derived on this basis, and the

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experimental results are obtained.

D. Phonon Transport Across Solid-Solid Interfaces

One experiment on the transport of nonequilibrium phonons across a solid-solid interface has been completed. Our findings greatly amplify our previous understanding of this phenomenon which has a number of technological implications. We find that, for phonon transport across an interface between a strong conducting film and a dispersive insulator, a strong spatial segragation occurs between high and low frequency phonons. Moreover, the high frequency component of the phonon distribution achieves a pseudo thermal equilibrium among themselves; the temperature of which decreases in time and in distance from the interface, until after many microseconds it reaches a common temperature with the low frequency component and finally true thermal equilibrium is obtained. Two efforts are underway to explain these results in terms of a theoretical basis. So far both methods yield an inconsistency with the data in regard to the relative (pseudo) temperature between the high and low frequency component of the phonon distribution. Further work on finding a theoretical basis is underway.

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II. Publications Resulting from Work Supported Under the

Contract

Defect Probes of Phonons in Insulating Crystals, W.E. Bron, "Defects in Insulating Crystals," Ed. by V.M. Tuchkevich and K.K. Shvarts, (Springer: Berlin) 1982, pg. 560.

Phonon Propagation by Quasidiffusion, W.E. Bron, Y.B. Levinson, and J.M. O'Connor, Phys. Rev. Lett. 49, 209 (1982).

Surface Requirements for Piezoelectric Generation of High Frequency Phonons, W.E. Bron, M. Rossinelli, Y.H. Bai, and F. Keilmann, Phys. Rev. B 27, 1370 (1983).

Lifetime of LO Phonons in GaP, J. Kuhl and W.E. Bron, Physica 117b/118b, 532 (1983).

Processes Involving Nonequilibrium Phonons in Solids, W.E. Bron, Optical and Acoustic Waves in Solids - Modern Topics, Ed. M. Borrisov (World Scientific: Singapore) 1983.

Temperature Dependence of Optical Phonon Lifetimes in GaP, J. Kuhl and W.E. Bron, submitted to Phys. Rev. Letters.

Phonon Generation, Transport, and Detection through Electronic States in Solids, W.E. Bron, Ed. W. Eisenmenger, "Nonequilibrium Phonons in Luminescent Crystals," to be published by North Holland.

Observation of a Quasidiffusive Phonon Propagation Mode, W.E. Bron, Y.B. Levinson, and J. O'Connor, to be published in the Proc. of the 4th Int. Conf. on Phonon Scattering.

Optical Phonon Dephasing and Depopulation Lifetimes, J. Kuhl, B.K. Rhee, and W.E. Bron, to be published in the Proc. of the 4th Int. Conf. on Phonon Scattering.

Heater Film Dynamics, Phonon Diffusion and Phonon Decay, T. Wilson, W.E. Bron, F.M. Lurie, and W. Schaich, to be published in the Proc. of the 4th Int. Conf. on Phonon Scattering.

III. Personnel Supported

- 1. Professor W.E. Bron, Principle Investigator.
- 2. Professor F.M. Lurie, Faculty Associate.
- 3. Dr. M. Rossinelli, Postdoctoral Associate.
- 4. T. Wilson, Graduate Student, Ph.D. expected by January 1984.
- 5. B.K. Rhee, Graduate Student, Ph.D. expected by September 1984.
- 6. D. Hurley, Undergradute Assistant.

