Frequency dependence of the scattering probability of nearly monochromatic phonons*

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The scattering probability of acoustic phonons propagating in SrF_2 containing 0.01-mole% Eu^{2+} has been investigated at three phonon frequencies. Superconducting fluorescer films of Sn, $Pb_{0.5}Tl_{0.5}$, and Pb are used to obtain, respectively, phonon distributions with narrow-band maxima at frequencies of 287, 407, and 670 GHz. Time-of-flight measurements of phonon pulses are taken with carefully monitored Al superconducting bolometers. The fraction of the time-integrated bolometer response which arises from scattered phonons is taken as a measure of the scattering probability. The experimental results are compared to a theoretical analysis based on the T-matrix formalism of the frequency dependence of the scattering probability from a mass disturbance with local force constant changes. The experimental results are in over-all agreement with theory.

I. INTRODUCTION

The scattering of phonons with frequencies greater than 100 GHz among each other or by lattice imperfections has traditionally been studied through thermal-conductivity experiments.[1] Although considerable information has been obtained by this method, the analysis of the frequency dependence of the scattering processes has been hampered by the fact that the phonons involved are not nearly monochromatic in frequency but have instead a blackbody distribution. More recently, a variety of schemes have been devised with which to generate phonon distributions with nearly monochromatic energies[2-6]; albeit such methods usually produce phonon populations considerably in excess of those used in thermal-conductivity experiments. For the most part, such phonon distributions have been used to date to demonstrate the existence[7] of resonant scattering centers in certain materials.[3,5,8,9]

This paper represents a first attempt to extract from these methods quantitative information on the frequency dependence of the relative scattering probability of nearly monochromatic phonons. The phonons propagate in a material containing point-lattice disturbances which give rise to a broad in-band scattering maximum at a frequency several times that of the phonon frequencies. The results are analyzed in terms of the T-matrix formalism[10] for in-band elastic scattering.

As will be seen, the experimental methods are not yet sufficiently developed such that a critical comparison between experiment and theory can be carried out. Nevertheless, the results do indicate that the experimentally determined frequency dependence of the scattering probability is in over-all agreement with that predicted by theory.

II. EXPERIMENTAL AND THEORETICAL BACKGROUND

Narayanamurti and Dynes[5,11] have shown that it is possible to generate phonon pulses with a dominant narrow-frequency component using what they call phonon fluorescence in superconducting films. The films are driven by a thin-film heater acting as a blackbody source of phonons. Phonons entering the superconductor with energies greater than the gap are converted to phonons with energies of the gap 2Δ through successive excitation, relaxation, and recombination of quasiparticles in the superconductor. Although the phonon distributions so obtained are not truly monochromatic, they do have a dominant component near the frequency $2\Delta/h$. (See Sec. IV for further discussion of these generators.)

The phonon pulses are propagated through single crystals of SrF_2 containing 0.01-mole % of Eu^{2+}. The choice of Eu^{2+} as a scattering center is fortuitous on many grounds. These grounds are: (i) Eu^{2+} ions substitute for Sr^{2+} ions maintaining thereby the cubic symmetry of the lattice. (ii) The lattice dynamics of SrF_2 and EuF_2 are well understood, as is the dynamical behavior of the SrF_2:Eu^{2+} system.[12-15] A detailed calculation of the scattering probability can therefore be carried out of this system. (iii) The pertinent physical parameters which characterize the dynamical properties of SrF_2 and EuF_2 are surprisingly similar.[16] This fact makes it possible to evaluate the effect of the Eu^{2+} disturbance in the SrF_2 lattice in terms of the difference in the dynamical parameters of the individual fluoride lattices.

The probability of phonons at three different frequencies being scattered by the Eu^{2+} disturbance is determined using time-of-flight measurements.[17] The amplitude of the phonon flux as a function of time is measured with a superconducting bolometer at the end of the crystal opposite to that of the phonon generator. The fraction of the time-integrated bolometer response which arises from scattered phonons, the so-called diffusive component, is taken as the measure of the scattering probability.

The theory of scattering of phonons by localized disturbances in solids has been developed by a number of authors. No attempt will be made here to review the subject. For present purposes it suffices to state only the final result. The expression obtained by Klein[10] for the scattering probability averaged over all phonons having a frequency ω is

$$-\langle\tau^{-1}\rangle_\omega = \frac{2pN}{\pi\rho^0(\omega)} \mathrm{Tr}[\mathrm{Im}\vec{t}(\omega^2 - i\epsilon)\,\mathrm{Im}\vec{g}(\omega^2 - i\epsilon)] \ . \quad (1)$$

In this expression, τ is an effective mean-free phonon lifetime which is assumed to exist in this formalism, p is the concentration of disturbance centers, N is the number of unit cells in the lattice, $\rho^0(\omega)$ is the (frequency) density of states, $\vec{t}$ is the T matrix, and $\vec{g}$ is the Green's-function matrix defined in the space of the coordinates of the disturbance. Specifically,

$$\rho^0(\omega) = \sum_{\vec{k}\lambda} \delta(\omega - \omega_{\vec{k}\lambda})/3rN, \quad (2)$$

$$g^{ij}(\vec{L}_\alpha, \vec{L}_\beta) = \frac{1}{N} \sum_{\vec{k}\lambda} \frac{\eta^i(\vec{L}_\alpha, \vec{k}\lambda)\eta^j(L_\beta, \vec{k}\lambda)}{\omega^2_{\vec{k}\lambda} - \omega^2 + i\epsilon} \ , \quad (3)$$

and

$$\vec{t} = \vec{\gamma}(\vec{1} + \vec{g}\vec{\gamma})^{-1} = (\vec{1} + \vec{\gamma}\vec{g})^{-1}\vec{\gamma}. \quad (4)$$

In Eqs. (2)–(4), $\omega_{\vec{k}\lambda}$ and $\eta(\vec{k}\lambda)$ are the eigenfrequency and eigenvector, respectively, of the vibrational mode of the unperturbed crystal with wave vector $\vec{k}$ and frequency branch λ; i and j refer to Cartesian components; L_α specifies the unit cell and location within the unit cell of a specific ion in the lattice; r is the number of ions in the unit cell; ϵ is an infinitesimally small number; $\vec{1}$ is the identity matrix; and $\vec{\gamma}$ is the so-called disturbance matrix defining the magnitude and coordinate space of the disturbance.

The Green's-function matrix is obtained directly from the eigenvectors of the pure lattice. For the eigenvectors, we use those obtained previously from the analysis, based on shell models, of vibronic and Raman spectra.[13,14] All pertinent parameters are given in Ref. 14. Experimental conditions in the present work limit the maximum phonon frequency to 670 GHz. In the SrF_2 lattice, phonons of this and lower frequency do not involve significant electronic shell polarization. That is, in the low-frequency regime, the lattice ions act as nearly nonpolarizable rigid ions. Accordingly, in the calculation of the Green's functions of Eq. (3), only the core eigenvectors (obtained, however, from the full shell-model treatment of the dynamical problem) are used.

III. EXPERIMENTAL AND THEORETICAL METHODS

The phonon generator is constructed as shown in Fig. 1. A thin film of 5000-Å thickness and 3-mm diameter of a superconductor, either Sn, $Pb_{0.5}Tl_{0.5}$, or Pb, is evaporated onto a newly polished end of a crystal of $SrF_2:Eu^{2+}$. A 3000-Å film of SiO of 8-mm diameter is evaporated over the superconductor, followed in turn by a constantan heater film and InSn alloy electrical contact tabs. The constantan heater film is 1×1 mm^2 in area and of a thickness such that the heater has a 50-Ω resistance at pumped helium temperatures. The Sn and Pb films are evaporated from resistively heated boats in the usual manner. In order to overcome problems in separation of the components of the PbTl alloy,[5] films of this material are produced by very rapid evaporation, to completion, of small chips of the alloy.

A voltage pulse of a few nanoseconds in duration is passed through the constantan heater. Considerable care must be exercised to match the voltage and duration of the pulse to the superconductor. In order to arrive at phonon pulses with comparable frequency distributions in each of the phonon fluorescer films the peak of the blackbody distribution in the constantan heater must bear an equivalent relationship to the gap frequency of each of the superconductors. If this is not done, the ratio of the nearly monochromatic phonon component to the total phonon flux emitted by the fluorescer will vary,[5,18] thereby severly complicating a comparative analysis of the scattering strengths. In this experiment, the magnitude of the voltage pulse was, accordingly, scaled such that in each case the maximum of the blackbody distribution nearly coincided with the corresponding gap ener-

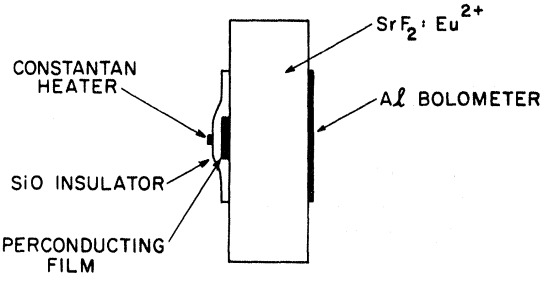

FIG. 1. Sketch of experimental sample with superconducting phonon fluorescer and bolometer.

gy.[5,18] For the present experimental situation, this means voltage pulses of the order of 7 V, 12.3 V, and 44.3 V across Sn, $Pb_{0.5}Tl_{0.5}$, and Pb films, respectively. For the case of Pb, it became necessary to reduce the pulse width to less than 35 nsec in order to prevent the superconductor from becoming normal. The procedure adopted to check this point will be discussed below.

SrF_2 crystals containing 0.01-mole % Eu^{2+} were obtained from Optovac[19] and cut and polished into cylindrical wafers 5 mm in radius and 3-mm thick. The phonon pulses are propagated along the 3-mm-long axis of the cylinder, which coincides with a [100] direction of the lattice. The surface equivalent to the cylinder wall is rough, which, together with the geometry of the sample, is designed to minimize sidewall phonon scattering.

An S-shaped aluminum thin-film bolometer is evaporated at the other end of the propagation path. The bolometer extends up to 4 mm from the axis such that phonons propagating along [100] and other directions are also intercepted by the bolometer. Typically, the Al film length to width ratio is 14:1, and the thickness ranges from 100 to 200 Å, corresponding to a dynamical range of several hundred ohms in the superconducting transition region.

The bolometers proved to be highly nonlinear, unless a set of precautions were undertaken. The procedure adopted was to require that variations of 10% of the following parameters did not result in a nonlinear change in amplitude over the entire time-resolved bolometer response. The parameters included the bolometer bias current (obtained from a constant-current source), the temperature, and the pulse-repetition frequency. After these values were set, the duraction of the voltage pulse through the heater film was varied. It was found, in order to satisfy the linearity criterion, that the pulse duration in the case of Pb fluorescers must be less than 35 nsec, but that longer durations are permissible in the case of Sn and the PbTl alloy.

Recovery of the time-resolved bolometer response is accomplished through normal boxcar integration. The main problem is to minimize electromagnetic cross talk between the generator and bolometer circuits. This is accomplished by careful placement of the generator and signal recovery lines and by constantly alternating the polarity of the generator driving voltage[20] within the time constant of the boxcar.

Evaluation of the frequency dependence of the scattering probability through Eq. (1) is carried out using the Green's-function matrices and disturbance matrices described by Lacina and Pershan.[21] The disturbance matrix includes the change in mass between the Eu^{2+} and Sr^{2+} ion, plus changes in force constants between the Eu^{2+} and the nearest fluorine ions, and between nearest-neighbor fluorine ions. The changes in the pertinent force constants are evaluated either from the methods described in Ref. 21, or directly from the differences in the microscopic parameters governing the lattice dynamics of the EuF_2 and SrF_2 lattices (See Table IV of Ref. 12 and Table III of Ref. 14). The resultant changes in the force constants, as defined by Ganesan and Srinivasan[22] and in terms of the notation of Lacina and Pershan, are (in units of dynes/cm^2) $\Delta\alpha_1 = -290$, $\Delta\alpha_3 = 680$, $\Delta\beta_1 = 300$, and $\Delta\beta_3 = -160$. The calculation was carried out for 1686 points in a $\frac{1}{48}$ part of the Brillouin zone. The result for the frequency dependence of the scattering probability is indicated in Fig. 2. Caution should be exercised in using this result for other than the purposes intended here. It is doubtful that the limitation to nonpolarizable scattering centers has meaning above 50 to 70 cm^{-1}. (The maximum phonon frequency used in this experiment is 22.6 cm^{-1}.) Nevertheless, it should be mentioned in passing that the strong resonance scattering in the 95-cm^{-1} region has been observed in the defect-induced far-ir absorption of this material.[23]

V. RESULTS AND DISCUSSION

The normalized time-resolved bolometer response, obtained for the three different phonon fluorescers, is shown in the solid lines of Figs. 3(a)–3(c). The first two sharp peaks represent the successive bolometer response to the fast longitudinal component and to a superposition of the two transverse components of the phonon pulse. The slowly decaying subsequent amplitude repre-

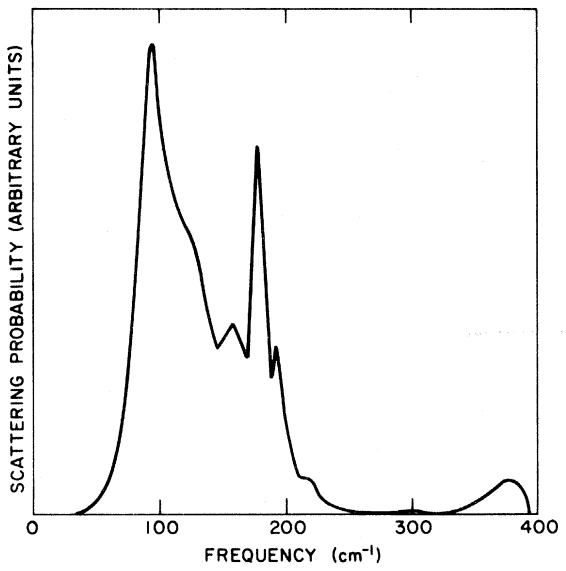

FIG. 2. Theoretically obtained frequency dependence of the scattering probability.

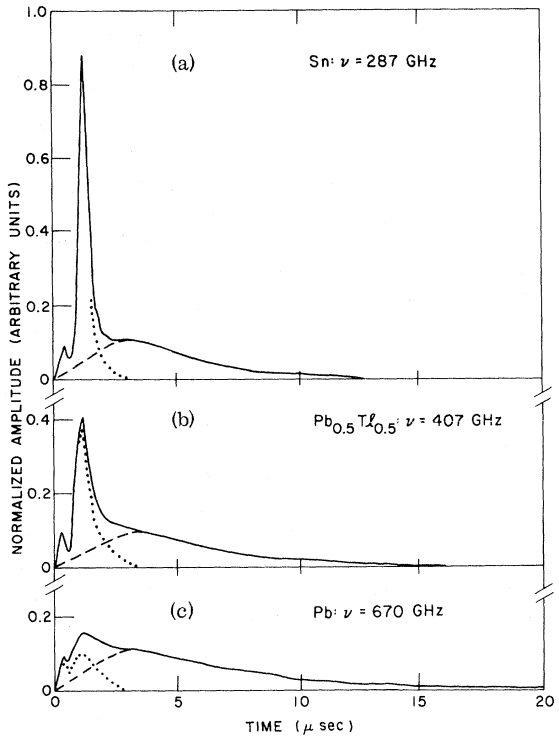

FIG. 3. Time-resolved bolometer response obtained from phonon pulses of three peak frequencies. The sample of SrF_2:Eu^{2+} is 3 mm thick and is held near 1.3 °K. The solid line is the normalized experimentally observed amplitude. The dashed line and the dotted line represent the analytical separation of the total response into the diffusive and ballistic components, respectively.

sents the diffusive component involving those phonons which have suffered one or more scattering events. The width of the individual ballistic components can be accounted for by the geometrical spread of phonon directions intercepted by the large bolometer film. The frequency dependence of the scattering process is quite readily observable.

The separation of the diffusive component from the ballistic component was accomplished analytically using the ramp (dashed line) geometry suggested by von Gutfeld[17] and the requirement that the half-width and trailing edge of the ballistic peaks be consistent among the three cases. The uncertainty inherent in the analytical method is one of the main contributors to the uncertainty in the amplitude of the frequency dependence of the scattering process. An even larger source of uncertainty is the magnitude of the miscellaneous scattering other than that caused by the Eu^{2+} ions. In order to determine this quantity, the experiments described above were repeated at the three frequencies using a "pure" sample of SrF_2 obtained from Optovac. The observed scattering represents 93% of that at 287 GHz using SrF_2 : Eu^{2+} crystals, and progessively much smaller fractions at the higher frequencies. The uncertainty in the Eu^{2+}-induced-scattering probability using Sn phonon fluorescers is, accordingly, higher than that in the other two fluorescers.

Uncertainties also exist as to the frequency span of the phonon distributions. Since the experimental arrangement does not include an element which measures this quantity directly, it is necessary to rely on auxiliary information. Dynes and Narayanamurti[5] have reported experimentally determined phonon distributions for Sn and $Pb_{0.5}Tl_{0.5}$ phonon fluorescers. Although their results may reflect in part the properties of the particular spectrographic element used, we adopt here as a basis for the uncertainty in phonon frequencies, the half width of the phonon distribution shown by them in Fig. 19 of Ref. 5. We assume further that the uncertainty in the case of Pb is comparable to that of Sn films, since the difficulty in producing homogeneous alloy films is not present in these cases. It should, however, be noted that some evidence exists that the upper bound on the frequencies of the phonons emitted from Sn films exceeds 670 GHz,[24] although the flux of such phonons is very small. The scattering probability, and the uncertainties, at the three available phonon frequencies are indicated by the data points of Fig. 4.

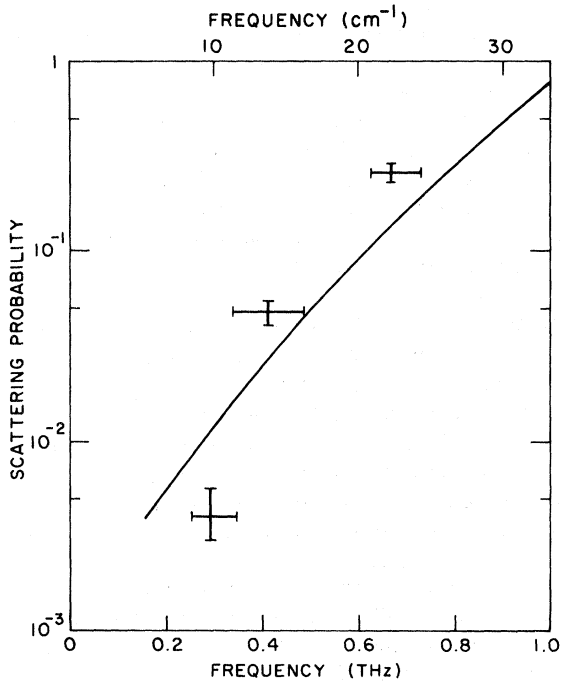

FIG. 4. Comparison of the experimental (data points) and theoretical (solid line) frequency dependence of the scattering probability.

For comparison, the calculated frequency dependence of the relative scattering probabilities is given by the solid line of Fig. 4. The amplitude of the calculated curve is scaled to yield the best overall agreement with the data.

A peculiarity of the results, as indicated in Fig. 2, deserves mention, This is the experimentally observed result that the area under the fast longitudinal component of the ballistic pulse appears to be relatively insensitive to the phonon frequency. The scattering probability does indeed vary inversely as the third power of the propagation velocity.[1] From the dispersion relationship of SrF_2, as given in Ref. 14, the velocity of longitudinal phonons propagating in a [100] direction is roughly twice that of transverse phonons. The scattering probability of longitudinal phonons is, accordingly, down by nearly a factor of 10. The data are, unfortunately, not accurate enough to check whether this factor accounts for the apparently small frequency dependence.

The relative intensity of the transverse to the longitudinal components of the ballistic signal is also of interest. Marked difference in the relative intensity of such peaks has been previously observed in heat pulse transport.[25,27] The effect has been explained in terms of differences between the two components in regard to acoustic mismatch at the generator-sample and sample-detector interfaces[25,26] and in regard to phonon-focusing effects[27] arising from the elastic anosotropy of the propagating medium. We have estimated the effects of acoustic mismatch using the methods of Ref. 26 and the effects of phonon focusing using the analytical method described by Maris.[27] These calculations predict the ratio of the longitudinal peak to the transverse peak to be 21 for Sn generators and 20 for Pb generators on samples of pure SrF_2 with [100] axis; these numbers should be compared to observed ratios of approximately 19 and 8, respectively. The origin of this discrepancy is not clear, although a likely contributor to the problem is the unfortunately large acceptance angle of the aluminum detector.

V. CONCLUSION

It is clear from Fig. 4 that the experimental methods are too crude at this stage to afford a critical comparison between experiment and the scattering theory based on the T-matrix formalism. Overall agreement on the relative frequency dependence of the scattering process is, however, indicated.

The experimental procedure can be improved in a number of ways. The most accessible is to change to the phonon generator described by Kinder,[4] which can be scanned continuously over the same frequency interval used here. The frequency spread probed by this method can be made narrower than that expected from the current technique. Moreover, a comparison of the results should shed some light on the as yet incompletely understood properties of these two methods of generating nearly monochromatic phonon pulses. Such experiments are currently underway and it is expected that they will be reported at a later time.

It is clear from Fig. 2 that extension of these experiments to the 50- to 70-cm^{-1} region should yield a stronger frequency dependence than that obtained at lower frequencies. This region is, however, relatively inaccessible to superconducting film generators of phonons. However, it does appear possible to generate phonons by optical pumping, with far-infrared lasers, directly into the defect induced absorption in the $SrF_2:Eu^{2+}$ system. It is also expected that the results of such experiments will be reported at a later time.

We conclude that although the experimental techniques are not yet completely developed, it does appear possible to probe the frequency dependence of the phonon scattering process using nearly monochromatic phonon pulses with frequencies in excess of 100 GHz. In the present experiment, such frequency dependence is readily observed and is in qualitative agreement with theory.

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[1]No attempt will be made here to review the large number of pertinent papers on thermal conductivity. For reviews of this field see, e.g., P. G. Klemens, in *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1958), Vol. 7; P. Carruthers, Rev. Mod. Phys. 33, 92 (1961).

[2]W. Eisenmenger and A. H. Dayem, Phys. Rev. Lett. 18, 125 (1967).

[3]K. F. Renk and J. Deisenhofer, Phys. Rev. Lett. 26, 746 (1971).
[4]H. Kinder, Phys. Rev. Lett. 28, 1564 (1972).
[5]R. C. Dynes and V. Narayanamurti, Phys. Rev. B 6, 143 (1972).
[6]E. H. Jacobsen, J. Phys. (Paris) 33, 25 (1972), (Suppl. 11–12).
[7]It is probably equally correct to state that the known resonant scattering in certain crystals have been used to demonstrate the various schemes to generate nearly monochromatic phonon distributions.
[8]H. Kinder, Z. Physik 262, 295 (1973).
[9]W. Forkel, M. Welte, and W. Eisenmenger, Phys. Rev. Lett. 31, 215 (1973).
[10]For an excellent review of the pertinent scattering formalism, see M. V. Klein, in *Physics of Color Centers*, edited by W. Beall Fowler (Academic, New York, 1968).
[11]V. Narayanamurti and R. C. Dynes, Phys. Rev. Lett. 27, 410 (1971).
[12]D. H. Kühner, H. V. Lauer, and W. E. Bron, Phys. Rev. B 5, 4112 (1972).
[13]L. L. Chase, D. H. Kühner, and W. E. Bron, Phys. Rev. B 7, 3892 (1973).
[14]D. H. Kühner and M. Wagner, Z. Physik 256, 22 (1972).
[15]W. E. Bron, Phys. Rev. B 11, 3951 (1975).
[16]See Table III of Rev. 12 for EuF_2. In this table, C_{11} should equal 12.40×10^{11} dyne/cm^2, $C_{12}=5.08\times10^{11}$ dyne/cm^3, $C_{44}=3.18\times10^{11}$ dyne/cm^2, $\omega(\Gamma, TO)=203$ cm^{-1}, and $\alpha_2=6.4\times10^{25}$ cm^{-3}, instead of the values indicated. For the corresponding values for SrF_2 see Table I of Ref. 14 in which R should read 2.89 Å.
[17]R. J. von Gutfeld, in *Physical Acoustics*, edited by W. P. Mason, (Academic, New York, 1968), Vol. V.
[18]Section II A 1 of Ref. 5 gives the pertinent relations between the maximum of the blackbody distribution, the temperature of the heater film, and the correspondingly required voltage.
[19]Optovac, Inc., North Brookfield, Mass.
[20]Since the sign of the electromagnetic crosstalk depends on the sign of the voltage but the phonon signal does not, it is possible to average out the crosstalk to a large extent by rapidly switching the polarity of the voltage within the integration time of the boxcar.
[21]W. B. Lacina and P. S. Pershan, Phys. Rev. B 1, (1970). However, the eigenvectors and eigenvalues from the polarizable shell model of Refs. 13 and 14 are used to obtain the unperturbed Green's functions.
[22]S. Ganasan and R. Srinivasan, Can. J. Phys. 40, 74 (1962).
[23]W. E. Bron (unpublished).
[24]I. Singer and W. E. Bron, Bull. Am. Phys. Soc. 20, 417 (1975).
[25]B. Taylor, H. J. Maris, and C. Elbaum, Phys. Rev. Lett. 23, 416 (1969).
[26]B. Taylor, H. J. Maris, and C. Elbaum, Phys. Rev. B 3, 1462 (1971).
[27]H. J. Maris, J. Acoust. Soc. Am. 50, 812 (1970).