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Frequency Distribution of Phonons in a Ballistic Heat Pulse Determined by Magnetic Resonance

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The scattering of ballistic heat pulses by resonant spins in $MgO:Fe^{2+}$, tuneable with a magnetic field, has been used to investigate the phonon frequency spectrum of the heat pulses at helium temperatures. A magnetic field of 60 kOe allowed phonons with frequencies up to 288 GHz to be studied. Good agreement was obtained with a Little-type calculation based on an acoustic-impedance model of interfacial thermal conduction. It is suggested that the lack of such agreement by most previous workers has often been due to inadequate preparation of surfaces.

I. INTRODUCTION

The method of heat pulses is now well established as a valuable technique for studying phonons in the frequency range 10–1000 GHz.[1] Its use has been considerably restricted, however, by uncertainty regarding the exact frequency spectrum of the phonons emitted into the specimen by the heat-pulse generator. Usually it has been assumed, following the theory of Little,[2] that

$$\rho(\nu)d\nu = \frac{2\pi k^4 A}{h^3}\left(\frac{\Gamma_l}{V_l^{\,2}} + \frac{\Gamma_{t1}+\Gamma_{t2}}{V_t^{\,2}}\right) \times\left(\frac{1}{e^{h\nu/k\theta}-1} - \frac{1}{e^{h\nu/kT}-1}\right)\nu^2 d\nu\ , \quad (1)$$

where $\rho(\nu)$ is the distribution of heat-pulse phonons as a function of frequency ν. In this expression, θ is the excitation temperature of the heat-pulse generator, T the ambient temperature of the specimen, A the contact area between generator and specimen, V_l and V_t the longitudinal and transverse acoustic velocities, and Γ_l, Γ_{t1}, and Γ_{t2} the phonon transmission coefficients for, respectively, longitudinal modes, transverse modes polarized in the plane of the interface, and transverse modes polarized normal to this plane. By the integration of (1) over all phonon frequencies an expression can be obtained which relates θ with P, the power dissipated in the heat-pulse generator

$$P = \left(\frac{2\pi^5}{15}\right)\frac{k^4}{h^3}A\left(\frac{\Gamma_l}{V_l^{\,2}} + \frac{\Gamma_{t1}+\Gamma_{t2}}{V_t^{\,2}}\right)(\theta^4 - T^4)\ . \quad (2)$$

This method of determining θ is unsatisfactory, however, because of the present lack of reliable values for Γ_l, Γ_{t1}, Γ_{t2} for any materials. A few thermal-conductivity measurements have been made of interfacial thermal conductance κ which is defined by $P=\kappa A\,(\theta - T)$ for $(\theta - T)\ll 1$, and therefore gives the proportionality constant in (2). Unfortunately, the experimental technique is a difficult one since it involves the extrapolation of temperature gradients on either side of the interface, and the data obtained have generally been inconsistent even amongst themselves.[3] In particular, they have not agreed with calculations based on the relative acoustic impedances of the two media, which might *a priori* be expected to determine the phonon-transmission probabilities, nor has any alternative theory been successful. Pulsed thermal-relaxation measurements on thin films have given data which lie somewhere between the acoustic-impedance values and calculations based on a perfect thermal-contact model.[4] By a radio-frequency mismatch technique Herth and Weis have measured the resistance and hence the temperature of a thin metallic film as a function of rf power.[5] With the exception of one result, to which we shall refer again later, their data also disagreed with the acoustic-impedance calculations.

A single previous attempt has been made to determine the heat-pulse phonon spectrum directly. Narayanamurti[6] used the resonant scattering (in zero magnetic field) of heat pulses by the ion V^{3+} in Al_2O_3 to measure their characteristic temperature. The value that he obtained differed from acoustic-impedance calculations by about 30%. In the present experiments, we have used a paramagnetic spin system as a variable-frequency phonon scatterer, tuneable by means of an external magnetic field, in order to determine the phonon-frequency distribution of ballistic heat pulses and thence to infer the interfacial thermal conductance. This is a particularly direct way of investigating the relation between θ and P, and significant clarification of interfacial thermal conduction may be expected.

II. EXPERIMENTAL TECHNIQUE

The spin system $MgO:Fe^{2+}$ was chosen for the experiment because its behaviour in a magnetic field can be represented by a very simple spin Hamiltonian, $\mathcal{H} = g\mu_B \vec{H}\cdot\vec{S}$ with $g = 3.43$ and $\vec{S} = 1$. In addition, its interaction with phonons is both strong and well understood as a result of many acoustic paramagnetic resonance and stressed-electron–spin-resonance measurements.[7] MgO is a cubic material and the specimen was cut as a $\langle 100\rangle$ right cylinder 6.7 mm in length and 6.0 mm in diameter, with the end faces polished optically flat. The heat-pulse generator was a thin film of constantan 680 Å thick and 4.2 mm^2 in area, that had been evaporated onto an MgO fact immediately after the latter had been polished and carefully cleaned. For this process the MgO specimen was left for several minutes immersed successively in Decon, acetone, trichloroethylene and ethyl alcohol in an ultrasonic bath at about 70 °C.

In order to generate heat pulses, the constantan film was excited by electrical pulses 200 nsec long at power levels between 10 and 500 mW. Its resistance was independent of magnetic field. A semiconducting avalanche bolometer of GaAs doped with 5×10^{17} cm^{-3} zinc detected the heat pulses as they reached the opposite face of the specimen.[8] The bolometer was 2.0 mm square and 0.1 mm thick, and was bonded to the MgO with a very thin film of epoxy resin. This type of semiconducting avalanche bolometer was much more sensitive than those reported previously,[8] and was capable of responding to generator power levels as low as 1 mW, corresponding to a generator temperature of 1.9K at the ambient temperature of the experiment 1.72 K. Such power levels were too low to cause saturation of the spin system $MgO:Fe^{2+}$. The sensitivity of the bolometer to incident phonons was independent of phonon frequency, and the method of its calibration against magnetic field has been described in detail in Ref. 8. Thus, a second crystal of pure Al_2O_3 containing no magnetic impurities was bonded to the rear face of the bolometer. Heat pulses passing through this material had an apparent variation with magnetic field due only to the magnetic-field-dependent sensitivity of the bolometer itself. The whole specimen assembly was immersed directly in liquid helium pumped to below its λ point.

Because of the finite sizes of generator and detector, the experiment involved all ballistic phonons travelling in directions less than 12° away from the $\langle 100\rangle$ axis. Only the transverse ballistic modes, degenerate for this cut of MgO, were observed as a result of phonon focussing.[9] The magnetic field was applied parallel to the axis of the specimen and thus to the predominant direction of heat-pulse propagation. In this configuration, the transverse phonons excited the $\Delta M = \pm 1$ transitions about two orders of magnitude more strongly than the $\Delta M = \pm 2$ line. Therefore, to a good approximation, a single band of phonons centered at $\nu = g\mu_B H/h$ was scattered from the heat pulse as it travelled through the MgO, with the bolometer detecting the unscattered residue. The maximum magnetic field that could be applied was 60 kOe corresponding to a frequency of 288 GHz. The first 200 nsec of the detected ballistic modes were sampled with a boxcar integrator and traced out as a function of magnetic field. Several such traces taken at different generator power levels are reproduced in Fig. 1.

Preliminary results from these investigations have been reported earlier.[10] The present experiments had an improved bolometer sensitivity of about 30 dB, and the maximum available magnetic field had been increased from 30 to 60 kOe. As a result, it was now possible to sweep the magnetic field through the condition $g\mu_B H \simeq 3k\theta$, corresponding, if the Little theory is correct, to the peak of

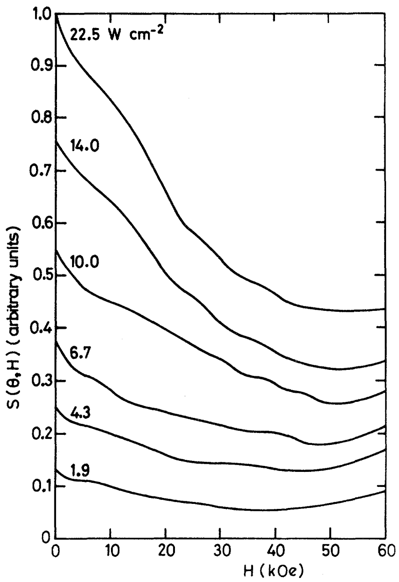

FIG. 1. Dependence of bolometer signal $S(\theta, H)$ on magnetic field H taken at different generator power levels.

the frequency spectrum $\rho(\nu)$.

III. INTERPRETATION OF DATA

The most important feature of the data was that the intensity of the heat pulses reaching the bolometer did indeed pass through a minimum, and furthermore, that the value of the magnetic field at which the minimum occurred increased with an increase in generator power. Both these observations supported qualitatively the conventional theory as summarized in expressions (1) and (2). At a fixed generator power level, a minimum in the heat-pulse intensity indicated a maximum in the number of phonons scattered by the spins, and therefore a maximum in the phonon distribution $\rho(\nu)$.

In addition to the broad minimum, however, there was also considerable structure which was believed to be due to Cr^{2+} present as an additional impurity in the MgO.[11] This ion undergoes a dynamic Jahn-Teller effect at helium temperatures, with the results, first that it is very strongly coupled to lattice vibrations, and second that it has several transitions lying below 1000 GHz in the frequency range of the heat pulses. The identification of the structure of Fig. 1 with particular Cr^{2+} transitions will not be discussed in the present communication. Although the scattering of phonons from the heat pulse by any single Cr^{2+} transition was small compared to that by the Fe^{2+} transitions, the total Cr^{2+} scattering did have the effect of reducing the transmitted heat pulse in zero magnetic field by an estimated 30%. The result was to increase the apparent absorption due to the Fe^{2+} transitions. In the absence of detailed knowledge on the Cr^{2+} transition probabilities, their effects were ignored, and the remainder of the analysis carried out on averaged smooth curves. The net result of this approximation seems merely to have been a slight increase in the scatter of the experimental points.

From the value of magnetic field at which a minimum of Fig. 1 occurred for a given generator power, it was possible to calculate quantitatively the temperature θ of the heat pulse. However, because of the large linewidth and asymmetric shape of the resonant absorption, it was not sufficient simply to assume that the maximum absorption of the heat pulse occurred exactly when ν_m (the peak frequency) and H_m (the magnetic field at the minimum of Fig. 1) satisfied the relation $h\nu_m = g\mu_B H_m$. Instead, it was necessary to evaluate completely the expression $S(\theta, H)$ for the total phonon energy reaching the bolometer as a function of θ and magnetic field H:

$$S(\theta, H) = C\int_0^\infty h\nu\rho(\nu)\exp[-\alpha(\nu) - \beta(\nu, H)]d\nu \ . \quad (3)$$

C defined the sensitivity of the bolometer and was eliminated by normalizing to $S(\theta, 0)$. Expression (1) was used for $\rho(\nu)$. The spin-phonon attenuation coefficient $\beta(\nu, H)$ was given by[12]

$$\beta(\nu, H) = (2\pi^2 l\nu/\rho v^3 \hbar)[(M_+^2/\epsilon^2)\, p_+(\nu, H)g_+(\nu, H) + (M_-^2/\epsilon^2)p_-(\nu, H)g_-(\nu, H)], \quad (4)$$

where the indices + and − refer, respectively, to the $\langle 1, 0| \longrightarrow |1, +1\rangle$ and $\langle 1, 0| \longrightarrow |1, -1\rangle$ transitions. The matrix elements $M_\pm$ of the spin-lattice Hamiltonian between the appropriate eigenstates were calculated to be[12]

$$M_\pm^2 = \tfrac{1}{2}\epsilon^2\,(\mu_B H F_{44} \pm G_{44})^2\,, \quad (5)$$

where F_{44} and G_{44} are the dipolar and quadrupolar magnetoelastic coefficients, respectively. Because of their degeneracy, transverse modes of all polarizations were assumed to be excited equally. For the evaluation of $M_\pm$ it was important to include quantitatively the dipolar as well the quadrupolar term. In most experiments using much lower magnetic fields the latter is completely dominant. The values of F_{44} and G_{44} were taken to be $+40$ unit strain^{-1},[13] and $+540$ cm^{-1} unit strain^{-1},[14] respectively.

The line shapes $g_\pm(\nu, H)$ of the transitions were obtained by extrapolation to high magnetic field and high frequency of results obtained in a separate (unpublished) acoustic-paramagnetic-resonance (APR) experiment. A Lorentzian shape with a half-width of 1.43 GHz at a frequency of 9.3 GHz and field of 2.0 kOe was found. Since the $MgO:Fe^{2+}$ resonances are inhomogeneously broadened by static-strain distributions in the MgO, the extrapolation could be carried out following directly the method of McMahon.[15] The atomic concentration of

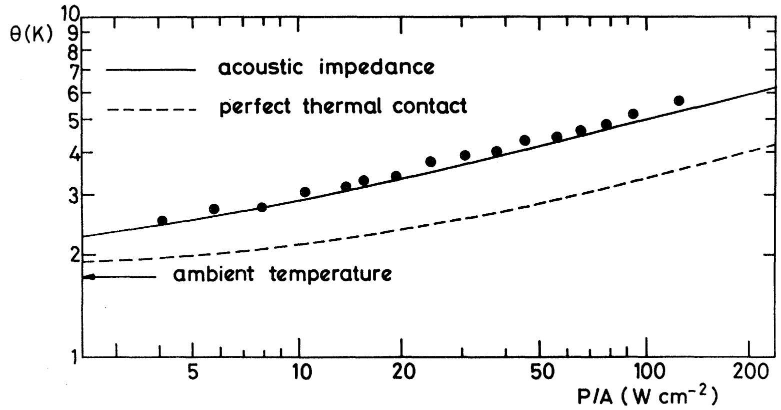

FIG. 2. Values of heat-pulse temperature θ obtained from the experimental data of Fig. 1 plotted against generator power per unit area P/A. Also shown are theoretical curves calculated using the acoustic-impedance and perfect-thermal-contact models.

Fe^{2+} ions was determined as 57 parts per million in the same APR experiment. The linewidths of the Fe^{2+} transitions were comparable in size to the ambient temperature so that it was necessary to calculate $p_{\pm}(\nu, H)$ the population differences, as frequency-dependent functions, after the method of McClintock *et al.*,[16] including both resonant $g_{\pm}(h\nu - g\mu_B H)$ and antiresonant $g_{\pm}(h\nu + g\mu_B H)$ terms.

Other symbols occurring in Eqs. (4) and (5) are l the length of the specimen, ρ the specimen density, v the appropriate acoustic velocity, and ϵ the magnitude of the acoustic strain. The only remaining term in expression (3) was $\alpha(\nu)$ which described the zero-field attenuation of phonons in the MgO, dependent on phonon frequency but not on magnetic field. An expression for $\alpha(\nu)$ was obtained from a preliminary investigation of the specimen performed in the regime of higher heat-pulse temperatures, $\theta \sim 15$ K.[10] Under these conditions $S(\theta, H)$ was much more sensitive to a variation in $\alpha(\nu)$ than to a change in θ and it was found that

$$\alpha(\nu) = (1.45 \pm 0.2) \times 10^{-47}\, \nu^{4\pm1} . \tag{6}$$

The form of this function suggested that the zero-field attenuation was due to mass-difference scattering by impurities in the MgO.[17] Although the measured concentration of Fe^{2+} could account for only about 15% of $\alpha(\nu)$, electron-spin resonance of the specimen indicated the presence of several other impurities (Fe^{3+}, Mn^{2+}, and Co^{2+}) in amounts similar to that of the Fe^{2+}.

Using a computer evaluation of $S(\theta, H)$ for the ranges of θ and H involved we were able to relate θ numerically to the magnetic field values at the minima of Fig. 1. Values of θ obtained by this method are plotted in Fig. 2 against generator power per unit area of contact.

IV. THERMAL CONDUCTION ACROSS AN INTERFACE

Also drawn in Fig. 2 are theoretical curves calculated for the two limiting cases of the perfect-thermal-contact and acoustic-impedance models of interfacial thermal conduction. For the acoustic-impedance calculation the two materials, constantan and MgO, were taken to be acoustically isotropic. An error of about 10% was introduced by this assumption. The four simultaneous equations with complex coefficients, relating the amplitudes of refracted and reflected phonons, were then set up for the three different polarizations of the incident phonons, longitudinal, transverse polarized in the plane of the interface, and transverse polarized perpendicular to this plane.[18] The equations were solved numerically to give the phonon-transmission probabilities $\alpha_k(i)$ for a phonon of polarization k and angle of incidence i. Finally, Γ_l, Γ_{t1}, and Γ_{t2} were calculated as

$$\Gamma_k = \int_0^{\pi/2} \alpha_k(i) \sin i \cos i \, di . \tag{7}$$

The values obtained were $\Gamma_l = 0.233 \pm 0.02$, $\Gamma_{t1} = 0.097 \pm 0.01$, and $\Gamma_{t2} = 0.062 \pm 0.006$. It is clear from Fig. 2 that the heat-pulse temperature θ can be described adequately by the acoustic-impedance calculation both as to its absolute magnitude and in its dependence on generator power.

The perfect-thermal-contact model describes the opposite hypothetical situation of all energy incident on the interface from either side being transmitted with 100% probability, regardless of angle of incidence. For this limit $\Gamma_l = \Gamma_{t1} = \Gamma_{t2} = 0.5$. It is difficult to see how this model can be valid when thermal conduction takes place predominantly by phonon transport. However, in a metallic film such as the heat-pulse generator the phonons are presumably in thermal equilibrium with the electrons, and it has been proposed[19] that energy may be transferred directly from the electrons in medium 1 to the phonons in medium 2, thus "shorting out" the phonon-phonon process. Recently, experimental evidence for an electron contribution to the thermal conduction across a metal-solid dielectric interface has been obtained by Wolfmeyer *et al.* and by Park and Narahara.[20]

Nevertheless, the results obtained in the present experiments suggest that less-fundamental effects associated particularly with inadequate preparation of surfaces may have been responsible for some of the inconsistent data obtained by previous workers. This view is supported by results of Herth and Weis[5] who obtained good agreement with acoustic-impedance calculations only on the one occasion that they cleaned the substrate and evaporated the metal film with extreme care. A small amount of oxide or impurity on one of the surfaces can change the acoustic matching very greatly at the short wavelengths possessed by thermal phonons. Other than Herth and Weis no previous workers have reported taking any special precautions to ensure clean surfaces.

A second point regarding the results of other workers is that most of their calculations for θ using the acoustic-impedance model have made graphical extrapolations of the curves given in Little's paper, instead of carrying out the more exact (and tedious) procedure summarized above. Again Herth and Weis[5] are the exceptions. Such extrapolations can give very inaccurate figures for materials having, as is usually the case, values of Poisson's ratio different from those corresponding exactly to Little's curves.

Finally, the author believes that the technique described in the present paper should give the most accurate data so far obtained in the study of thermal conduction across an interface, since all adjustable parameters have been eliminated for the comparison between theory and experiment.

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