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Phonon Scattering in Condensed Matter

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Fourth International Conference
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August 22-26, 1983

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Preface

This volume contains the proceedings of the Fourth International Conference on Phonon Scattering in Condensed Matter held from August 22-26, 1983 at the University of Stuttgart. The preceding conferences were organized at Saint Maxime and Paris in 1972, at the University of Nottingham in 1975, and at the Brown University Providence/Rhode Island in 1979.

The Stuttgart conference, like the preceding conferences, was mainly concerned with "propagating" high-frequency acoustic phonons, mechanical waves and heat up to the lattice limiting frequency. Lattice dynamics, optical phonons, phase transitions, etc., were included as far as they are involved in acoustical phonon scattering, propagation and generation. In this context the conference covered all aspects of acoustical phonon physics, especially generation of phonons, propagation, scattering and detection. Since acoustic phonons participate in most energy-transfer processes in solids and liquids, the field of interest is growing rapidly. Therefore exciting new developments of acoustic phonon physics could be presented at the Stuttgart conference as well as important progress with respect to well-known problems, as, for example, the Kapitza resistance.

Two hundred and six scientists from 21 countries attended the conference. Thirteen invited papers and 105 contributed papers, with 34 as posters, were presented. The discussions are included in this volume.

A discussion session on large wave vector phonons was organized and chaired by V. Narayanamurti. A discussion session on phonon scattering at interfaces was organized and chaired by R.O. Pohl.

The conference was supported and sponsored by the Deutsche Forschungsgemeinschaft, the International Union of Pure and Applied Physics, the Land Baden Württemberg, the University of Stuttgart, the Deutsche Physikalische Gesellschaft and the E. Leitz Company, Wetzlar. The support and sponsorship of these organizations is gratefully acknowledged.

We extend our thanks to the Rektor of the University of Stuttgart, Prof.Dr.rer.nat. H. Zwicker, who opened the conference and gave a reception for the delegates.

We should like to thank all members of the international advisory committee for their extremely helpful suggestions and recommendations.

We thank our colleagues of the organization committee K. Dransfeld, E. Sigmund and M. Wagner for close and fruitful cooperation. Special thanks are due to our colleague H.-J. Bauer for his advice and assistance in adapting our data handling system to the needs of the conference. In particular we thank Mrs. R. Mann, Mrs. I. Poljak and Mrs. G. Untereiner as well as all our co-workers and students for their invaluable assistance and help during all stages of the conference.

Stuttgart, December 1983 *W. Eisenmenger · K. Laßmann · S. Döttinger*

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Part I

Acoustic Phonon Spectroscopy

Chairmen:
W. Dietsche K. Dransfeld T. Ishiguro G. A. Northrop
J. P. Wolfe A. F. G. Wyatt

Crossing Effects in Phonon Scattering

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Resonant scattering from a phonon current can occur at the transition frequencies ν_i associated with the electronic or motional energy levels of impurity ions or centres present in the crystal. If the concentration of centres is 'small' the scattering will only be significant within a narrow frequency bandwidth Δ [1] which may be much less ($\lesssim$ 1 GHz) than that of the phonon current ($\sim 2kT$ for a thermal current or $\sim$ 40 GHz at 1K). Under these conditions the frequency spectrum of the phonon current has sharp holes burned in it as shown in fig. 1(a). In many cases these can be moved to and fro by applying an external perturbation such as a magnetic field. Sharp features in the total scattering occur when two of these holes cross ($\nu_i = \nu_j$) or in some cases anticross and these provide spectroscopic information which can be of quite high precision and resolution.

There are three main effects [2], frequency, level and anticrossing.

1. Frequency Crossing

This effect only requires that 2 transition frequencies become equal. It was first observed and accounted for qualitatively by BERMAN et al. [3] using the heuristic model illustrated in fig. 1(b) which assumes that when resolved, the 2 resonant processes completely block the 2 rectangular conduction channels shown shaded. However, when they are crossed only one channel is blocked so the total conduction rises giving a signal $\Delta K/K_0 \sim$ channel conduction/total conduction and width $\sim$ channel width. In practice of course a channel is never entirely blocked. Its conduction depends on $\tau(\nu)$, the phonon relaxation time and $\tau(\nu) = (\tau_B^{-1} + \tau_1^{-1} + \tau_2^{-1})^{-1}$, where τ_B^{-1}, τ_1^{-1} and τ_2^{-1} are respectively the background and two resonant scattering rates. Expansion of $\tau(\nu)$ contains functions of the product $\tau_1^{-1}\tau_2^{-1}$ which is essentially zero at all frequencies when the two processes are resolved, but becomes non-zero for $\nu \sim \nu_1 = \nu_2$ when the processes are crossed [Fig 2]. So the crossing signal is a consequence of the non-linear dependence of $\tau(\nu)$ on the scattering rates.

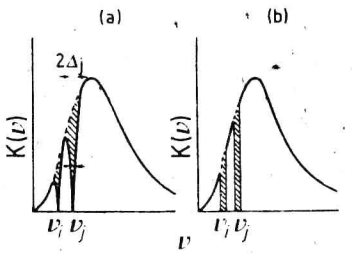


Figure 1

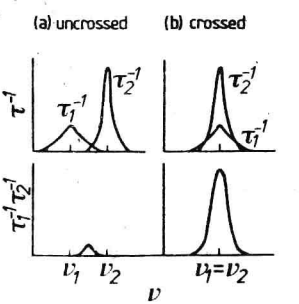


Figure 2

At low temperatures and in a Debye model the conductivity differs from that of the pure crystal by

$$\Delta K = K_o - K = CT^3 \int_o^\infty \frac{x^4 e^x}{(e^x-1)^2} \left[\tau_B - \tau(\nu) \right] d\nu \text{ where } x = h\nu/kT.$$

In general the crossing signal $\Delta W/W_o \tilde{-} -\Delta K/K_o$ ($W = 1/K$) can be computed from τ_1^{-1} and τ_2^{-1} by calculating ΔK as a function of the separation of the 2 frequencies in the region of the crossing frequency ν_o. However, when the holes have widths $\ll kT$, we can simplify the calculation since

$$\Delta K \tilde{-} CT^3 \left[\frac{x^4 e^x}{(e^x-1)^2} \right]_{\nu_o} \int_o^\infty \left[\tau_B - \tau(\nu) \right] d\nu.$$ Indeed in some limits the integral can be solved analytically for both Lorentzian and Gaussian line shapes giving simple expressions for the form of the signal [4]. So the size and shape of frequency crossing signals can provide information on the resonant scattering rates and hence the spin-phonon coupling constants or, if these are known, on the ionic concentrations. Their positions can provide spectroscopic information such as the spin-Hamiltonian parameters. Although this discussion refers to steady heat currents, similar analysis of course applies to heat pulses.

An example of a frequency crossing signal is shown in fig. 4 at low resolution. The signal shows the change in temperature difference along a sample as the field is swept and is usually displayed at $\Delta W/W_o$. The system is V^{3+} in Al_2O_3 whose levels for B||c axis correspond to those of fig. 3 and give rise to a crossing at a field $B = D/3g_{11}\beta$. The crossing moves to higher fields when these are moved away from the c axis, and D and $g_\perp$ can be obtained rather accurately from the straight line plot of B^{-2} against $\cos^2\theta$ (θ is the angle to the c axis) if g_{11} is known from EPR which is very often the case in such systems [5].

The upper 2 levels of fig. 3 are in fact each split into 8 by hyperfine interaction, as shown in fig. [3]. This gives rise to the structure shown in fig. [5], [7]. Figure 6 shows the differential of this signal obtained by applying a small modulation field to the sample [8].

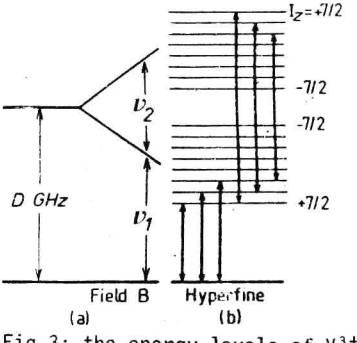


Fig. 3: the energy levels of V^{3+} in Al_2O_3. If we neglect hyperfine splitting ($\sim 10^{-3}$D), $\nu_1 = D - g_{11}\beta B$ and $\nu_2 = 2g_{11}\beta B$ so $\nu_1 = \nu_2$ when $B = D/3g_{11}\beta$

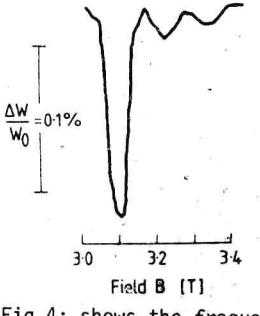
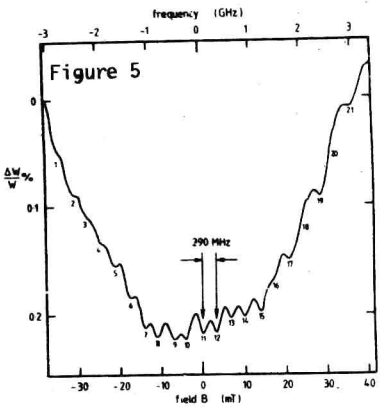
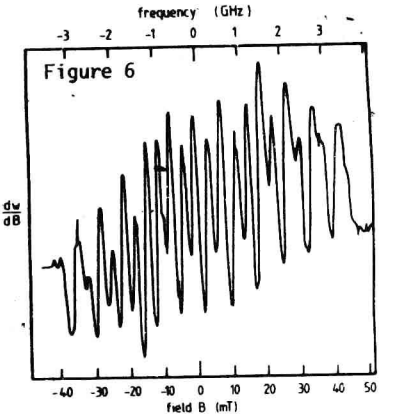


Fig 4: shows the frequency crossing signal of fig 3 displayed at low resolution. The 2 signals at higher fields are due to V^{3+} pairs present at a concentration $\sim 10^{-2}$ppm [6]





In this case and many others that have been reported the 2 scattering processes were both from the same type of ion but crossing signals have also been seen when the processes are from 2 ions in inequivalent sites [3], an ion and an ion pair [6] and 2 entirely different ionic species [9], [10]. So, as WALTON first stressed [9] one can use a known ion as a probe to investigate another and the fact recently demonstrated that they do not have to be in the same part of the sample opens up the scope of the technique.

2. Frequency Crossing between spatially separated ions

This has been demonstrated [11] using an Al_2O_3 bicrystal doped with Fe^{2+} ions in one half and V^{3+} in the other [Fig. 7]. Several crossings occur between the transition frequencies of the two ions and fig. 7 shows two of these (lines C and D). The increase in their size when heat injection is switched from H_V to H_{Fe} shows that the Fe^{2+} holes in the phonon current are carried across the interface; the V^{3+} ions have little effect on this current when tuned to a hole. This technique seems capable of being developed in various ways and we hope to use it to study ions in epitaxial and diffused layers, electrons in inversion layers, etc. by crossing their frequencies with those of probe ions in the bulk crystal [12].

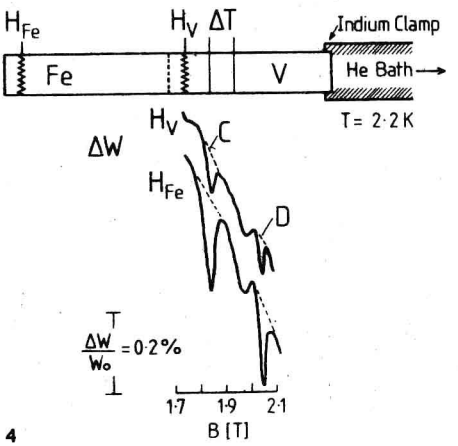


Figure 7: shows frequency crossing signals from the Fe/V Al_2O_3 bicrystal shown in the upper part of the figure. The signals ΔW are minima in the temperature difference on the V-doped half. The signals are ∿ 3 times larger when heat is injected at H_{Fe} rather than at H_V in the control experiment when the signals are due to traces of Fe^{2+} in the V half

3. Level Crossing

The effects that should be observable here using phonons are analogous to the HANLE effect seen in light scattering in 1924 [13] and first explained by BREIT in 1933 [14]. In recent years it has been used as a technique for high resolution spectroscopy [15]. Suppose an ion has 2 crossing levels with energies $h\nu_1$ and $h\nu_2$ above a singlet ground state and that the upper 2 levels can be made to cross by a magnetic field (fig.8(a)) making $\nu_1 = \nu_2$. If white light is incident on the ion, a photomultiplier can be used to measure the total intensity I_T of the components at ν_1 and ν_2 scattered into a particular small solid angle. Now if $|\nu_1 - \nu_2| \gg \gamma$ (γ is their combined line width), $I_T = I(\nu_1) + I(\nu_2)$ but if $|\nu_1 - \nu_2| \lesssim \gamma$, interference takes place in the emission process and $I_T = |A(\nu_1) + A(\nu_2)|^2 = I_1 + I_2$ plus an interference term. So the photomultiplier records a signal of width γ at the crossing point. There seems no reason to suppose that similar effects should not occur with phonons although so far as I know, no experiments have been reported. It might be possible to see them in thermal conduction which would certainly be affected by changes in the angular distribution of the scattered phonons that occur at a level crossing. However, at the concentrations used in phonon experiments so far, these changes would be accompanied by frequency crossing effects and while the resulting signal would certainly differ from a pure frequency crossing signal [16], it is not clear whether these differences could be identified. So although it would seem that it would be interesting to explore these effects in this way, experiments looking more directly at the angular distribution would seem to be more promising.

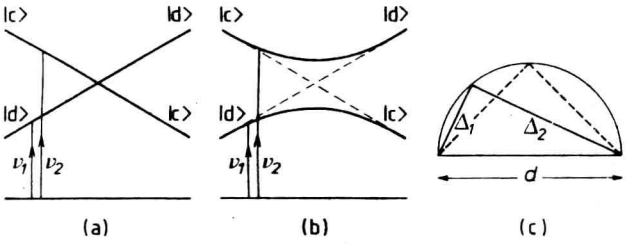


Figure 8

A complication in level crossing experiments comes from the effect of random strain which may prevent levels from approaching to within their linewidths (anticrossing).

4. Anticrossing

If there is coupling between the 2 approaching levels of fig.8(a), they will repel each other or anticross as shown in fig.8(b). So if the holes in the heat current at ν_1 and ν_2 are narrower than their nearest separation, they can never overlap and no frequency crossing signal can occur. However, there can still be another sort of signal because of the state mixing that occurs at the anticrossing. This changes the widths Δ_1, Δ_2 of each of the holes while keeping $\Delta_1^2 + \Delta_2^2$ constant. The additional thermal resistance caused by the existence of the holes α $(\Delta_1 + \Delta_2)$ which has a maximum at the centre of the anticrossing when the 2 hole widths become equal $(\Delta_1 = \Delta_2)$. This can be seen in Fig 8(c) where $\Delta_1^2 + \Delta_2^2 = d^2$ (d = diameter of semicircle)