

future ultrasonic work at high magnetic fields are obvious. First, a resolution of the magnetic field direction to within  $0.1^\circ$  is necessary in order to take accurate high-field data in ultrapure single crystals, and, in the analysis of such data, a  $B^{-2}$  saturation rate should not be assumed in advance for metals with open Fermi surfaces. Second, the possibility of drastically different contributions from various parts of Fermi surfaces supporting open orbits should be taken into consideration when trying to extract electron relaxation times from the high-field attenuation.

## ACKNOWLEDGMENTS

We wish to express our appreciation to Dr. P. R. Antoniewicz for many helpful discussions and to Dr. L. T. Wood for the use of his computer program based on the model copper Fermi surface developed by Dr. M. R. Halse. We are also grateful to Dr. A. F. Clark for making the high-purity copper available, to Dr. E. Mielczarek for the use of the potassium crystals, and to S. J. Chang and J. P. Masson for their assistance with the experiments.

<sup>†</sup>Work supported by a grant from the National Science Foundation.

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## Displaced Distribution Functions for Three-Phonon Umklapp Processes

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(Received 12 November 1969)

It is shown that three-phonon umklapp processes tend to displaced Planck distributions that are characterized by the reciprocal-lattice vectors of the crystal.

Since the phonon frequency and the phonon group velocity are periodic functions of the phonon wave vector, it is a convenient representation to describe the wave vector in the reduced-zone scheme. The distinction between the three-phonon normal and umklapp processes occurs only in this scheme. It is well known that in the presence of an equilibrium thermal gradient, the normal processes tend to a displaced Planck distribution.<sup>1-4</sup> This is because

these processes conserve the phonon wave vector (or phonon momentum), and the distribution to which they tend should be consistent with this condition. We put some arguments in the following to emphasize that similar distribution functions also exist for the three-phonon umklapp processes.

If a phonon is represented by  $(\vec{q}j)$ , where  $\vec{q}$  is the phonon wave vector and  $j$  the polarization index, then the three-phonon processes are described by

$$(\tilde{q}_1 j) + (\tilde{q}_2 j) = (\tilde{q}_3 j). \quad (1)$$

The occurrence of this process demands the fulfilment of the following conditions<sup>5</sup>:

$$x_1 + x_2 = x_3 \quad (2)$$

and

$$\tilde{q}_1 + \tilde{q}_2 = \tilde{q}_3 + \tilde{g}, \quad (3)$$

where  $x$  is the reduced phonon frequency, i. e.,  $x = \hbar\omega/kT$ ,  $\omega$  being the phonon frequency. Here  $\tilde{g}$  can be a null vector or one of the reciprocal-lattice vectors of the crystal. The three-phonon normal processes correspond to  $\tilde{g} = \tilde{0}$ ;  $\tilde{g} \neq \tilde{0}$  describes the umklapp processes.

It is generally thought that the umklapp processes destroy the wave vector completely, but this is not correct, as is clear from Eq. (3). Since  $\tilde{q}$ 's are confined to the first Brillouin zone, it is obvious that there can be only a limited number of reciprocal-lattice vectors that can be involved in the umklapp processes. Furthermore, a given three-phonon process can correspond to one and only one reciprocal-lattice vector (one which brings  $\tilde{q}_1 + \tilde{q}_2$  to the first Brillouin zone). It is, therefore, possible to divide the umklapp processes into different classes, a class being characterized by a definite value of the reciprocal-lattice vector. Let us call  $u(\tilde{g})$  processes all those three-phonon umklapp processes which involve a given reciprocal vector  $\tilde{g}$ . For the  $u(\tilde{g})$  processes, we have from Eq. (3)

$$(\tilde{q}_1 - \tilde{g}) + (\tilde{q}_2 - \tilde{g}) = \tilde{q}_3 - \tilde{g}. \quad (4)$$

Equation (4) clearly shows that the  $u(\tilde{g})$  processes conserve the vector  $(\tilde{q} - \tilde{g})$ , and therefore they should tend to a distribution which is consistent with this condition. One can find this distribution function in the same way as is used in the case of normal processes. This distribution, denoted by  $N_{u(\tilde{g})}$ , should be given by

$$\{\exp[\hbar\omega/kT + \tilde{\lambda}(\tilde{g}) \cdot (\tilde{q} - \tilde{g})] - 1\}^{-1}, \quad (5)$$

where  $\tilde{\lambda}(\tilde{g})$  is a constant vector which should be proportional to the thermal gradient in the lowest-order approximation. It can be evaluated in the same way as  $\tilde{\lambda}$  in the case of normal processes (see Ref. 1).

We justify here the division of the umklapp processes into the  $u(\tilde{g})$  processes. The division of the three-phonon processes into normal and umklapp processes is made on the grounds that normal processes satisfy a condition (wave-vector conservation) which the umklapp processes do not satisfy. It is therefore possible to study the normal processes separately. We emphasize that it is not sufficient to stop the division at this stage. One should recognize the distinction between different umklapp processes. There are some umklapp processes

which satisfy Eq. (4) and others that do not. It is therefore justified to study the  $u(\tilde{g})$  processes separately. In this picture, we may call  $u(\tilde{0})$  the normal processes because they can be thought to be those for which  $\tilde{g}$  equals the null vector.

We can also obtain the displaced distribution (5) in a different way. We start with the net transition probability with the collision event (1). This probability is proportional to

$$[N_1 N_2 (N_3 + 1) - (N_1 + 1)(N_2 + 1) N_3]$$

or  $N_1 N_2 N_3 F$ , where

$$F = N_3^{-1} - N_1^{-1} - N_2^{-1} - N_1^{-1} N_2^{-1} = F_{12,3}, \quad (6)$$

$N = N(\tilde{q} j)$  being the phonon distribution. We use a trial function

$$N^{-1} = e^{x \tilde{\lambda} \cdot (\tilde{q} \cdot \tilde{G})} - 1 \quad (7)$$

to minimize the value of  $F$ . It can be seen that because of Eqs. (2) and (3),

$$F = e^{x \tilde{\lambda} \cdot (\tilde{q}_3 \cdot \tilde{G})} [1 - e^{\tilde{\lambda} \cdot (\tilde{g} \cdot \tilde{G})}]. \quad (8)$$

It can be seen that if one selects  $\tilde{G}$  equal to  $-\tilde{g}$ ,  $F$  can be made to vanish identically for any given three-phonon process involving  $\tilde{g}$ . Consider an hypothetical case in which there are only  $u(\tilde{g})$  processes; these collision events will redistribute phonons in various modes till the transition rates become minimum. In other words, they tend to minimize the deviation of  $F$  from zero, that is, these processes tend to the drifted (displaced) distribution (7), with  $\tilde{G} = -\tilde{g}$ , which is distribution (5).

Consider the Boltzmann-transport equation in the presence of normal processes, which can be written in the form

$$\tilde{\nabla} \cdot \tilde{\nabla} T \frac{dN_i}{dT} = \sum C N_1 N_2 N_3 [F_{12,3} - \frac{1}{2} F_{23,1}], \quad (9)$$

where  $F_{23,1}$  corresponds to  $(\tilde{q}_1 j_1) = (\tilde{q}_2 j_2) + (\tilde{q}_3 j_3)$  and  $C$  is a function of the phonons involved and of anharmonicity, etc. Summation is to be performed over  $\tilde{q}_2 j_2$  and  $\tilde{q}_3 j_3$ . It is obvious that solution to Eq. (9) can be found in the form given by (7) with  $\tilde{G} = \tilde{0}$  in the limit  $\tilde{\nabla} T \rightarrow 0$ . This solution corresponds to a rigid flow of phonons.<sup>2,6</sup> This suggests that normal processes alone cannot obstruct the heat flow. In fact, had it been possible to have only  $u(\tilde{g})$  processes, one would have obtained a similar result for them also.

It needs mentioning that the distribution given by (5) is not actually what the phonons attain. The actual phonon distribution  $N$  is decided by all the scattering processes that can take place in a crystal and the thermal gradient applied. One might say that the distribution given by expression (5) is not justified since it might lead to negative occupation numbers. It is not at all significant if this happens, since  $N_{u(\tilde{g})}$  is not the distribution that is attained by

the phonon system. We can remove this confusion by calling  $N_{u(\vec{g})}$  a constant such that the strength of the  $u(\vec{g})$  processes is proportional to the difference of  $N$  (actual distribution) and  $N_{u(\vec{g})}$ . Such a constant, even if it is negative, does no harm in any way provided a solution to a real problem starting with this concept does not suffer from any physical inconsistency. It should be noted that all three-phonon processes take place side by side in a real crystal, since they are due to the same anharmonicity.

A study of the umklapp processes in the way we have made is important in the sense that it gives a unified treatment of the three-phonon processes. We distinguish different processes by the reciprocal-lattice vectors (including null vector) they involve. Assigning the displaced distribution function of the form (5), makes it possible to determine the individual contribution of the  $u(\vec{g})$  processes. If we assume that  $\vec{\lambda}(\vec{g})$  is parallel to  $\pm \vec{\nabla} T$ , we can arrive at certain interesting results. Consider  $u(\vec{g})$  processes alone, and let  $\vec{g}$  be perpendicular to  $\vec{\nabla} T$ ; then these processes do not change the phonon momentum in the direction of  $-\vec{\nabla} T$ , and therefore should not be expected to obstruct heat flow like the normal processes. They should offer resistance to heat flow indirectly by redistributing phonons in different modes like the latter. The result is not changed if we consider  $u(\vec{g})$  and  $u(-\vec{g})$  processes

simultaneously, and therefore the conclusion that these processes should be studied simultaneously, and that they together tend to Planck's distribution is not valid. Thus, umklapp processes by themselves do not necessarily contribute to thermal resistance. In fact, the contribution of different  $u(\vec{g})$  processes will depend on the orientation of  $\vec{g}$  and the value of  $\lambda(\vec{g})$  that is decided by the thermal gradient.

Truly speaking, the notion that the wave-vector conservation necessarily means conservation of the heat flow is approximately correct, since the heat flow is associated with the group velocity of phonons which is not always constant in magnitude and parallel to the phonon wave vector. Very often it can be antiparallel to the phonon wave vector.<sup>7</sup>

If one starts with the Boltzmann equation as usual and makes use of the displaced-distribution functions, one can find an expression for the thermal conductivity depending explicitly on the reciprocal-lattice vectors, or in other words, on the symmetry of the crystal. This means that besides other factors that decide anisotropy in the thermal conductivity of a crystal, the three-phonon processes also contribute to it through the displaced distributions. The above discussion thus makes it possible to study the effect of different  $u(\vec{g})$  processes in a completely revised way.

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