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ON AN APPARENT RESTRICTION IN THE INELASTIC He SCATTERING FROM CLOSE PACKED METAL SURFACES: A COMMENT

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It is demonstrated experimentally that there is no fundamental restriction in detecting phonons travelling in either direction with respect to the parallel component of the incident He beam on close packed metal surfaces.

The inelastic He scattering data obtained so far by Toennies' group [1] and by Lambert et al. [2] from metal surfaces exhibited a surprising behavior: both the created and the annihilated Rayleigh wave phonons seemed to travel in the same direction, namely in the direction opposite to the projection on the surface of the incident He atom velocity. Apparently, phonons travelling "with" the incident He atoms could neither be created nor annihilated. Each time-of-flight (TOF) spectrum showed correspondingly at most *one* Rayleigh phonon peak; the interaction being anyhow restricted to the first Brillouin zone due to the very low periodic corrugation of the close packed metallic surfaces used in the experiments. In view of the fixed scattering angle ϑ_d in both experimental set ups ($\vartheta_d = 90°$ in ref. [1] and $\vartheta_d = 120°$ in ref. [2]) phonon creation was seen only for $\vartheta_i < \vartheta_d/2$ while annihilation only for $\vartheta_i > \vartheta_d/2$ where ϑ_i is the incident angle of the He beam with respect to the surface normal. This behavior was at variance with the inelastic He scattering data from alkali halides [3].

Lambert et al. [2] plotted their phonon dispersion data (fig. 2 in ref. [2]) in a very appropriate manner to emphasize the behavior described above. The data points measured in the experiments done so far on metal surfaces were lying exclusively in the quadrants I and III (see also fig. 1 below, where the same kind of plotting is used). Cautiously, Toennies [1] confined himself to a plain statement of the apparent facts ("the conditions are such that essentially only phonons with $\omega > 0$, $Q < 0$ and $\omega < 0$, $Q > 0$ are observed for $\vartheta_i > 45°$ and $< 45°$, respectively"), without mentioning that this behavior might imply a definite propensity: He interacts only with phonons travelling in a particular direction. However, Lambert et al. [2] sought an explanation for a behavior which seemed to be an interesting physical effect. They analyzed first the influence of "kinematical focusing" on the phonon scattering cross section. The analysis led them to a result exactly opposite to the data: the dominant processes should appear in the quadrants II and IV. Then, based on arguments from Meyer's paper [4], they suggested that dynamical effects should overcompensate the influence of the kinematical focusing. In particular, Lambert et al. concluded that the above behavior (data points either in quadrant I or III and none in II and IV) is observed always when $v/c \sin\vartheta_i \gtrsim 0.4$, where v and c are the velocities of the incident He atom and of the sound for the surface phonon branch of interest, respectively.

We show below that the behavior observed so far originates in simple kinematics which impedes under certain conditions the measurement of data points in the quadrants II and IV; i.e. that there is no propensity related to the travelling direction of the phonons with respect to the incident direction of the He beam. We demonstrate that by an appropriate choice of the experimental conditions data points in these quadrants can be obtained; it is even possible to measure TOF curves in which Rayleigh phonon annihilation *and* creation peaks are present simultaneously and this, according to Lambert et al., under the most "unfavorable" condition $v/c \sin\vartheta_i \approx 1.2$.

In fig. 1 scan-curves for $E_i = 18$ meV and for several incident angles ϑ_i are plotted as dashed lines under the geometrical condition of our experiment ($\vartheta_d = 90°$ as in ref. [1]). Also plotted is the dispersion relation for the Rayleigh phonons on the Pt(111) surface in the $\overline{\Gamma M}$ direction calculated by Black et al. [5]. Phonon creation or annihilation can be observed at the intersections of the dispersion curves with the scan curve which corresponds to the experimental parameters during the TOF scan. It is obvious that the dispersion curve lying in the quadrants I and III can be intersected over a broad range of incident angles. The plotted triangles correspond to the peaks seen in our TOF spectra for $E_i = 18$ meV measured in the $\overline{\Gamma M}$ direction of the Pt(111) surface. The agreement with the dispersion curve calculated by Black et al. (model I in ref. [5]) is good.

The only problem with detecting the creation or annihilation of phonons travelling "with" the He beam, is to find the appropriate conditions to intersect the dispersion curve lying in the quadrants II and IV. In view of the general shape of the scan-curves, the range of the incident angles ϑ_i for which the dispersion curve is intersected extends only a few degrees around $\vartheta_d/2 = 45°$, i.e. around the very intense specular beam. The presence of this very intense beam hinders TOF measurements to be made at ϑ_i very close to $\vartheta_d/2$. In order to intersect the dispersion curve in the quadrants II and IV without coming too close to $\vartheta_d/2$ a more appropriate shape of the scan curve, adapted to the shape of the dispersion curve, has to be found. We have done this by taking e.g. $E_i = 33.4$ meV. Two of these scan curves which lead to data points

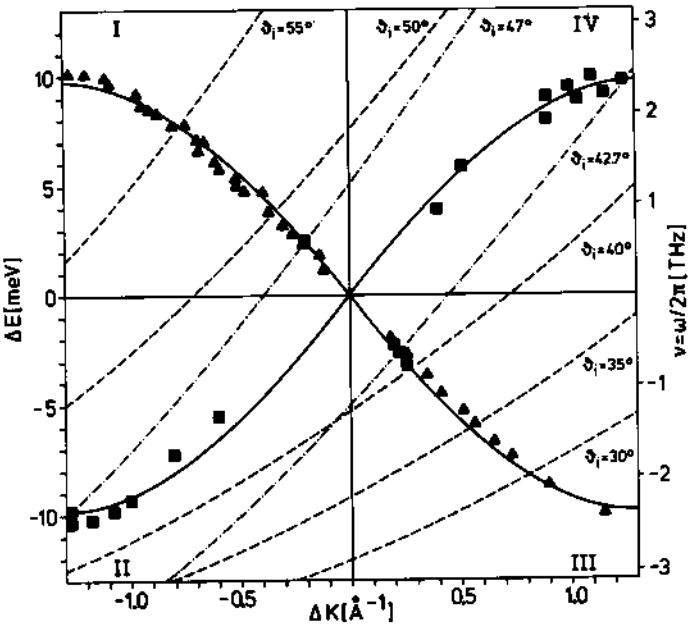

Fig. 1. Energy change ΔE versus surface component of the wave vector change ΔK of He atoms scattered in the $\vartheta_d = 90°$ geometry from a Pt(111) surface in the $\overline{\Gamma M}$ direction. Dashed and dash-dotted lines are, respectively, scan curves for different ϑ_i and for $E_i = 18$ and 33.4 meV He incident energy. Triangles and squares correspond to unambiguous Rayleigh phonon peaks in TOF spectra for $E_i = 18$ and 33.4 meV, respectively. The solid lines are the Rayleigh phonon dispersion curves calculated in ref. [5].

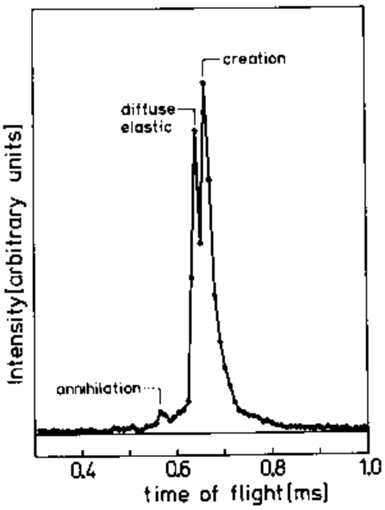

Fig. 2. Measured TOF spectrum for $E_i = 33.4$ meV and $\vartheta_i = 42.7°$ (see corresponding scan curve in fig. 1).

in the quadrants II and IV are also shown in fig. 1 (dash-dotted). The plotted squares which represent the peaks seen in the corresponding TOF spectra demonstrate that also creation and annihilation of phonons travelling "with" the He beam can be detected.

In fig. 2, one of these TOF spectra obtained with $E_i = 33.4$ meV and $\vartheta_i = 42.7°$ is shown (the flight path between chopper and detector is 79 cm). Besides the diffuse elastic peak both a phonon annihilation and a phonon creation peak are clearly visible. From the corresponding scan curve in fig. 1 we deduce that the annihilated phonon was travelling "with" the He beam while the created phonon "against" it. The large intensity ratio of the two peaks is in fair agreement with calculations of Celli et al. [6], the annihilation peak being near the Brillouin zone boundary.

In conclusion, we have demonstrated experimentally that there is no principal restriction in detecting phonons travelling in either direction on close packed metal surfaces and thus under appropriate conditions phonon creation and annihilation can be detected simultaneously also on these surfaces.

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