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INTERACTION OF 25-keV ELECTRONS WITH LATTICE VIBRATIONS IN LiF. EXPERIMENTAL EVIDENCE FOR SURFACE MODES OF LATTICE VIBRATION*

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In two former papers^{1,2} it has been stated that monoenergetic³ fast electrons interacting with molecules excite infrared active fundamental vibrations with high intensity. Consequently, strong interaction of fast electrons with optical lattice vibration should be expected in alkali-halide crystals. In these solids, however, as is well known, electromagnetic waves excite transverse vibrations ω_T of the lattice, whereas a charged particle excites the longitudinal modes ω_L . The maximum of light absorption and the most probable energy loss appear at different energies.

Figure 1 represents (a) the real and the imaginary part of the dielectric constant $\epsilon(\omega) = \epsilon_1(\omega)$ $+i\epsilon_2(\omega)$, (b) the reflection spectrum (reststrahl band, and (c) the energy-loss function $-\text{Im}[1/\epsilon(\omega)]$ according to the dielectric theory⁴ of an infinite medium, in lithium fluoride. The absorption spectrum, which is essentially proportional to ϵ_2 , shows a rather sharp peak at $\hbar \omega_T = 0.039$ eV. The reststrahl band extends over a wide range from 0.03 to 0.08 eV. The energy-loss function calculated from the optical constants exhibits a peak at $\hbar \omega_L = 0.081$ eV, where ϵ_1 vanishes, in accordance with the Lyddane-Sachs-Teller relation.⁵

The high-resolution technique recently developed³ enables us now to separate those electrons which have lost such small amounts of energy from the unaffected electrons at 25-keV primary energy. In contrast to the energy-loss function, the measured energy-loss spectrum of a 400-Å LiF foil (Fig. 1) shows a broad band between 0.03 and 0.11 eV with a maximum at 0.047 eV. The spectrum is corrected by subtracting the slope of the no-loss line.¹ The excitation of lattice vibrations takes place with largest probability at an excitation energy between those of the transversal and longitudinal modes. The energy-loss spectra depend slightly on foil thickness (Fig. 2). The maximum shifts from 0.042 to 0.050 eV when the foil thickness increases

from 240 to 700 Å.

In addition to these energy losses, energy gains of the primary electrons could also be observed in the spectrum (Fig. 2) changing their position in the same way. The energy gains disappear when the specimen is cooled down to the temperature of liquid air. Since processes leading to energy loss and energy gain have equal probabilities, the intensity ratio of energy gain and energy loss is proportional to the occupation probability of the state, which is a function of the temperature of the specimen.

The discrepancies between the computed and measured energy-loss spectra in Fig. 1 are caused by the fact that we are dealing with a finite medium. On the assumption that the excitation probability of lattice vibrations ω_L is



FIG. 1. Dielectric constant $\epsilon = \epsilon_1 + i\epsilon_2$, reststrahl band *R*, energy-loss function $-\text{Im}1/\epsilon$, and experimental energy-loss spectrum (normalized) of LiF on carbon substratum, $E_0 = 25 \text{ keV}$, $\theta \le 1 \times 10^{-4}$. Optical data taken from M. Gottlieb, J. Opt. Soc. Am. <u>50</u>, 343 (1960).



FIG. 2. Energy gains and energy losses of 25-keV electrons in LiF for different foil thicknesses *D* on carbon substratum. $\theta \ge 10^{-4}$. $T = 300^{\circ}$ K.

given by the dipole approximation,⁶ the polarization waves excited in the LiF foil have wavelengths between 700 and 70 000 Å in the range of scattering angles $0 \le \theta \le 1 \times 10^{-4}$ which are accepted by the analyzer. These wavelengths are much longer than the thickness of the foils. As Fröhlich⁷ already pointed out for the case of a sphere with a radius small compared with the wavelength, the transversal and the longitudinal vibrations should be degenerate. The joint excitation energy should be nearly midway between $\hbar \omega_T$ and $\hbar \omega_T$.

To explain our results we slightly generalize



FIG. 3. Dispersion of the "tangential" (ω_{-}) and "normal" (ω_{+}) surface modes of lattice vibration. The experimental points have been obtained assuming that the scattering angle θ' essential for the position of the maxima is given by $\theta'' = 1.2\hbar\omega/E_0$, i.e., the angle at half-width of the angular distribution. k_0 is the wave number of the primary electrons.

the dielectric theory of surface plasma oscillations in metal foils given by Ritchie⁸ and apply the dielectric formalism to the excitation of surface-lattice vibrations by inelastic electron scattering. The theory now is closely related to that of vibration modes in an ionic crystal slab recently developed by Fuchs and Kliewer.⁹ Thus the observed energy-loss spectra with maxima between 0.042 and 0.050 eV can be interpreted as "tangential" surface-lattice vibrations ω_{-} . The thickness dependence of the position of the maximum is due to the dispersion of this surface mode (Fig. 3). The excitation probability of the "normal" surface mode ω_+ and of the longitudinal vibration ω_L of the bulk medium seems to be so weak that it does not show up in the spectrum as a pronounced maximum.

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SUPERCONDUCTING MIXED-STATE-STRUCTURE DETERMINATION IN VANADIUM BY NUCLEAR MAGNETIC RESONANCE AND RELAXATION

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It is now generally accepted that within type-II superconductors in the mixed state there can exist an ordered array of lines, each surrounded by a vortex-like circulation of current.¹ As a result the magnetic field in the metal varies periodically in space. Neutron-diffraction studies in niobium have given direct evidence for this structure,² showing that the vortices are arranged on a triangular lattice whose lattice constant (typically 100 to 1000 Å) is determined by the same flux quantization condition per vortex as is found for superconducting cylinders.³ We report here line-shape measurements⁴ which strongly suggest that the vortex structure is also triangular in the mixed state of vanadium^{5,6}; and relaxation-time (T_1) measurements⁷ for nuclei both near and far from vortex lines.

Both measurements use the field-cycling technique⁸ shown in Fig. 1, in which a 10-kG field is first applied to the normal sample for a time long compared to T_1 ; then the field is lowered, in 30 msec, to some value H_e such that the sample becomes superconducting. The field is kept at H_e for a time τ , during which the spin magnetization relaxes with a time characteristic of the mixed state. Finally the field is rapidly increased to 6 kG and the nuclear spin magnetization measured by immediately observing the nmr signal (which is proportional to the magnetization) using standard techniques. The cycle is repeated many times with varying τ , and frequently the signal decays exponentially with τ towards its $\tau = \infty$ value, the time constant being T_1 .

Except near H_{C2} and T_c , the relaxation data⁹ are consistent with the assumption that far from vortex lines spins relax at the rate predicted for type-I superconductors by the BCS theory¹ [$T_1 \approx (0.8 \pm 0.4) \exp(1.75T_c/T)$ sec]; and spins within a coherence distance of a vortex relax at roughly the same rate as in the normal metal.¹⁰ Thus, at low T and H_e , nonexponential decays are observed having a long ex-

ponential tail with the BCS T_1 quoted above, and having a rapid initial decay associated with spins close to vortices. The space average of T_1^{-1} can also be obtained from the initial slope of the decay, or more accurately by applying a 100-Hz magnetic field of 10 G in the direction of H_{ρ} . This ac field presumably moves the vortex structure about relative to the spins, so that each spin relaxes at the average rate for the entire structure, and a shortened, purely exponential decay is observed. The average rate $\langle T_1^{-1} \rangle$ is proportional to T as in the normal state, and is also proportional to the density of vortices $[\langle T_1^{-1} \rangle \simeq \frac{1}{2} T_{1n}^{-1} B / H_{c2}$, where T_{1n} is the normal-state¹⁰ relaxation time at the same temperature. These observations are consistent with theoretical ideas of Caroli and co-workers¹¹ and with other kinds of experiments.^{5,12}

Just below T_c the characteristic drop in T_1 relative to the normal state found in other superconductors¹ is not seen in our samples for $H_e = 0$. There is a drop in T_1 for $H_e \approx 50$ G just below T_c , relative to the normal state at the same field. The absence of this decrease in zero-field T_1 may be a real effect, but it could also be the result of trapped flux since, in the normal state, T_1 increases by a factor of about



FIG. 1. Field sequence used to measure T_1 and line shape. The 100-Hz audio field sometimes used to move the vortex structure is shown dotted. The dotted magnetization and signal curves show what happens when power is absorbed by the nuclei from the probe field.