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Electron-Spectrometric Study of Amorphous Germanium and Silicon in the Two-Phonon Region

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The energy-loss spectra of 25-keV electrons transmitted through thin amorphous Ge and Si films have been measured in the range of very small energy losses between 25 and 400 meV. The resolution was 4 and 6 meV. The main intensity in the spectra is located at low energy and is caused by "defect-induced" two phonon excitation.

In a series of former papers[1,2] monoenergetic fast electrons (25–35 keV primary energy) were shown to interact strongly with optical phonons and surface phonons[3] in ionic crystal films. The strength of interaction of fast electrons appeared to be quite similar to that of electromagnetic radiation, which is strongly absorbed by ionic crystals at the frequencies of the transverse optical vibrations because of the large dipole moment connected with the vibrating lattice.

In the present paper the interaction of electrons with thin films of amorphous germanium and silicon is investigated. Since these targets consist of atoms of the same sort, light can couple with lattice vibrations only via transition moments of higher orders, and weak absorption due to two-phonon excitation is observed instead. For example, the maximum absorption coefficient of LiF is[4] $K = 40\,000$ cm^{-1} which is to be compared with $K = 30$ cm^{-1} in the two-phonon region of Ge.[5] Two-phonon excitations by fast electrons, therefore, are expected to be very weak, and energy losses corresponding to these processes would be observed only under great difficulties. Fortunately, as it turned out, these difficulties were not that serious because the intensity of the corresponding energy losses can be enhanced to a certain extent by the distorted structure of the films.

The highly resolved electron energy-loss spectra of Ge and Si (Figs. 1 and 2) were obtained by means of a Wien filter spectrometer.[6] The energy of the electrons was 25 keV, the energy resolution 4 and 6 meV. The self-supporting films (thickness between 150 and 650 Å) have been prepared by evaporation on to a freshly cleaved NaCl crystal. Electron diffraction shows the amorphous structure of the films. During handling the films were exposed to open air so that some oxidation of the films occurs.

The intensity of the energy losses within the phonon region (indicated in Figs. 1 and 2) depends on the conditions during the preparation of the film, that is on the residual gas pressure and on the rate of evaporation, if the pressure is not too low. The phonon intensity increases with decreasing rate of evaporation and with increasing pressure. The same behavior is found for the electric resistance ρ_s of the films.[7] One reason for this dependence is that during the evaporation the material is contaminated by residual gas atoms, mainly oxygen. Secondly, the microstructure of the films will depend on the conditions during evaporation, in the sense that the distortion of the Ge tetrahedra, their mutual order, and the density of voids vary. Hence it is concluded that the noticeable increase in intensity of the two-phonon energy losses is due to "defect-induced" two-phonon excitation. This effect is quite analogous to defect-induced one-phonon excitation discussed by Dawber and Elliot.[8] Accordingly the two-phonon intensity is particularly weak for the films evaporated in ultrahigh vacuum or for heat-treated polycrystalline films. Simultaneously the underlying background continuum indicated in Fig. 1 increases. The posi-

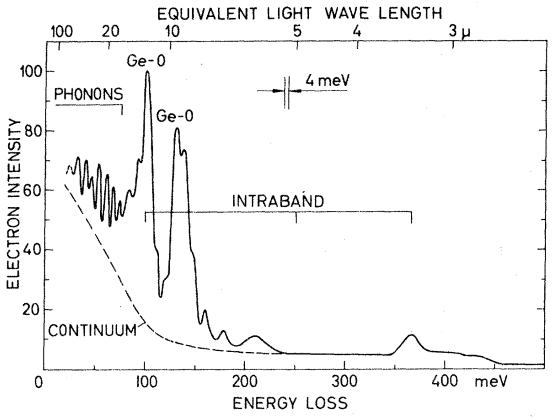

FIG. 1. Energy-loss spectrum of germanium. Thickness of the film 250 Å, $\rho_s = 2.5\times10^3$ Ω cm; primary electron energy 25 keV, and scattering angle $\theta < 1.1\times10^{-4}$.

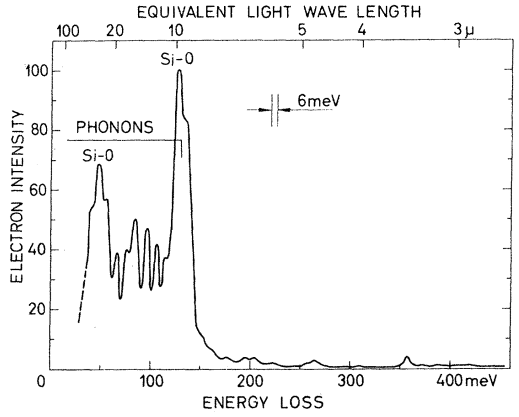

FIG. 2. Energy-loss spectrum of silicon. Thickness of the film 200 Å, $\rho_s = 1.5\times10^8$ Ω cm; primary electron energy 25 keV, and scattering angle $\theta < 1.1\times10^{-4}$.

tion of the energy-loss peaks in the two-phonon region agrees very closely with the maxima of the two-phonon density of states[9] of the bulk media (see Table I).

In order to estimate the contribution of surface modes and bulk modes to the electron intensity within the energy-loss spectra, the dielectric theory of inelastic electron scattering in thin solid films (see, for example, Geiger[10]) has been applied to obtain a theoretical energy-loss spectrum. The complex dielectric constant which is required for evaluating the scattering formula was derived from optical measurements.[5] In doing so a constant value of the real part of the dielectric constant[11] $\epsilon_1(\mathrm{Ge}) = 16$ and $\epsilon_1(\mathrm{Si}) = 14.4$ was assumed, which is justified[9] in the case of weak absorption ($K\lambda/4\pi \ll 1$). The calculation leads to the following results: The shape of the energy-loss spectrum is essentially determined by the spectral behavior of the imaginary part of the dielectric constant, ϵ_2. The reason for this is that ϵ_1 is approximately constant and $\epsilon_2 \ll \epsilon_1$. The shape of the spectrum is the same for bulk modes and for surface modes. In the range of film thicknesses investigated the main contribution to the electron intensity of the spectra in Figs. 1 and 2 comes from excitation of surface modes. The electron intensity due to bulk modes amounts to 10% and less.

From a different point of view, which is based on the incoherent neutron-scattering cross section, DeWames and Hall[12] have pointed out that the shape of an electron energy-loss spectrum is closely related to the phonon density of states. This result is in remarkably good agreement with our experiment (Table I). The theory neglects surface effects. In the present case, however, as has been shown before, the spectral behavior of the surface and bulk modes is identical.

The experimental results presented here do not contradict Ibach's work[13] on energy losses of low-energy electrons reflected from silicon single-crystal surfaces. In these measurements only a single energy loss at 55 meV has been found. The numerous losses listed in Table I, which reflect the two-phonon density of states, could be observed in our measurement because the structure of the film was highly distorted.

Several energy losses in Figs. 1 and 2 must be interpreted as due to Ge-O and Si-O vibrations. They are located at 100 and 135 meV in the Ge spectrum and at 49 and 128 meV for the Si spectrum; the latter values are consistent with the re-

TABLE I. Energy losses E (averaged) and maxima of the two-phonon density of states[a] E_{ph} of germanium and silicon, in meV.

Ge		Si	
E	E_{ph}	E	E_{ph}
32	33	40	42
40	39	49[b]	
46	44	58	58
53	53	66	66
61	60	76	75
68	65	86	86
74	72	96	93
		104	105
		113	115
		128[b]	126

[a] See Ref. 9.

[b] Mixed with and covered by Si-O vibration peaks.

sults in Ref. 13.

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Left-Right Asymmetry in the Scattering of Electrons by Magnetic Impurities, and a Hall Effect

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Left-right asymmetry is expected in the scattering of electrons by magnetic impurities in metals; we present a simple calculation of the asymmetry in a virtual-bound-state model. When a magnetic field lines up the impurity moments, this left-right asymmetry should generate an extraordinary Hall effect proportional to the impurity concentration. This effect appears to occur in many magnetic alloys (e.g., Ni-, Cu-, and Au-based alloys).

It has been known[1] for a long time that the scattering of fast (few MeV) spin-polarized electrons by nuclei exhibits a left-right asymmetry which is induced by spin-orbit coupling. When the scattering potential depends on the spin only through the spin-orbit term, the asymmetry becomes inverted for electrons of opposite spin; and so, for unpolarized electrons, the effect of the electrons with opposite spin balances the asymmetry. On the other hand, this balance cannot occur and an asymmetry remains when the scattering potential —even without the spin-orbit term—is different for each spin direction. This is the case for the potential of magnetic impurities in a metal. We shall show that resonant scattering on magnetic-impurity levels may exhibit an observable asymmetry when the spin-orbit coupling is not much smaller than the width of the impurity virtual bound state. This left-right asymmetry should generate a Hall effect: Smit,[2] Luttinger,[3] and Kondorskii, Cheremushkina, and Kurbaniyazov[4] have already predicted this contribution—proportional to the impurity concentration—to the extraordinary Hall effect of ferromagnetics.

First, we will give a simple calculation of the asymmetry in a virtual-bound-state model of the scattering magnetic impurity. We consider an impurity having only a spin moment,[5] like Mn or Fe in Au. We assume also that the impurity is placed in a magnetic field strong enough to prevent scattering through spin-flip of the impurity moment ($\mu H \gg kT$) or Kondo scattering ($\mu H \gg kT_K$) so a Hartree-Fock approximation can be used to describe the impurity scattering; in the Friedel-Anderson model[6,7] we have then to consider spin-up and spin-down virtual bound states (vbs) in the conduction band near the Fermi level (see Fig. 1; we assume for simplicity that we deal only with d vbs).

In the usual Friedel-Anderson picture, the spin-orbit coupling is neglected (and the crystal field also) and the scattering of a free conduction electron with spin σ and energy E is mainly determined by the only phase shift of the $l=2$ par-