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[11]The presence of charge-density-wave-like distortions means that diffuse scattering can occur both from soft phonons and from elementary excitations of the charge-density waves or phasons. Scattering from phasons has been considered by Overhauser (Ref. 7) and W. L. McMillan (to be published) and it has been stated that its chief effect is to produce strong diffuse scattering in the immediate vicinity of the charge-density-wave Bragg-scattering peak similar to the diffuse scattering that is normally described in terms of a Debye-Waller factor. The extended diffuse scattering normal to the *c* axis which is the subject of this paper does not immediately appear to be of this type. The fact that the transition from the normal phase to the incommensurate phase is a second-order transition means that the existence of a small static distortion at a particular point in the reciprocal lattice can be accompanied by softened phonons in the vicinity of that point. These softened modes can of course produce strong diffuse scattering. We would like to remind the reader that the recently published work of D. E. Moncton, J. D. Axe, and F. J. DiSalvo [Phys. Rev. Lett. 34, 734 (1975)] was on the 2*H* polytypes of these materials and as they point out, the transitions to the incommensurate and commensurate phases in the 1*T* and the 2*H* systems are considerably different.

Giant Quantum Oscillations in the Attenuation of Rayleigh Waves in Gallium

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Giant quantum oscillations in the attenuation of Rayleigh waves with the magnetic field normal to the metal surface are reported. These oscillations have a spikelike character. The experiments are made at frequencies up to 120 MHz and temperatures down to 0.4 K. The line shape is studied and compared with that of the giant quantum oscillations in the attenuation of accoustic bulk waves in longitudinal magnetic field.

I report the first observation of giant quantum oscillations (GQO) in the attenuation of Rayleigh waves in a metal in a transverse magnetic field. A theoretical study of this effect has been recently reported by Grishin and Kaner.[1] This phenomenon is different from the GQO in the attenuation of acoustic bulk waves which were predicted by Gurevitch, Skobov, and Firsov[2] and observed in gallium by Shapira and Lax.[3] These GQO arise from the resonant absorption of the sound wave by the electrons which move along the magnetic field with the sound velocity. So, they do not exist when the acoustic wave vector is perpendicular to the magnetic field. In this Letter, it is shown that the attenuation of Rayleigh waves in a magnetic field normal to the metal surface experiences oscillations with a spikelike character. In the case of Rayleigh waves, the spatial inhomogeneity of the deformation field smears out the conservation law of the electron-momentum projection on the magnetic field and the electrons can absorb energy from the Rayleigh wave in a resonant process.[1]

I now describe briefly the experimental procedure.[4] The single crystal is made with very pure gallium in a plastic mold which has two polished faces.[5] The flat surfaces of the gallium crystal are normal to the $\vec{b}$ axis.[6] A ZnS film about 6000 Å thick is evaporated on the crystal. This piezoelectric film acts as an electromechanical transducer. Two metallic combs are evaporated on the ZnS film. The comb teeth are 20 μm wide and a distance of 40 μm from each other. Each comb consists of 40 teeth and is a distance of 6 mm from the other one. A rf pulse (about 0.5 μsec duration) is applied between one comb and the sample (the metallic sample is grounded). A Rayleigh wave is generated on the metal surface if the frequency of the rf pulse is such that the tooth spacing equals a whole number of Rayleigh wavelengths. The acoustic surface wave propagates normal to the comb teeth and reaches the second comb (which is parallel to the first one) after a certain time. At this time a rf pulse is observed on the second comb. No reflection of the acoustic pulse on the combs is observed, so there are no successive echoes (as in the case of bulk waves) and it is not possible to measure the absolute value of the attenuation. The experiments are made at low temperatures and high magnetic fields. The magnetic field is produced by a superconducting magnet and the low temperatures are obtained with a He^3 cryostat (the sample is in the helium bath).

The GQO with spikelike character appear at magnetic fields above 20 kOe. Figure 1 shows a typical recorder tracing of the change in the attenuation of Rayleigh waves as a function of magnetic field intensity. The peaks are periodic in inverse magnetic field. The measured period is 29.7×10^{-7} Oe^{-1} and is equal to the period of the GQO in the attenuation of longitudinal bulk waves in longitudinal magnetic field.[3]

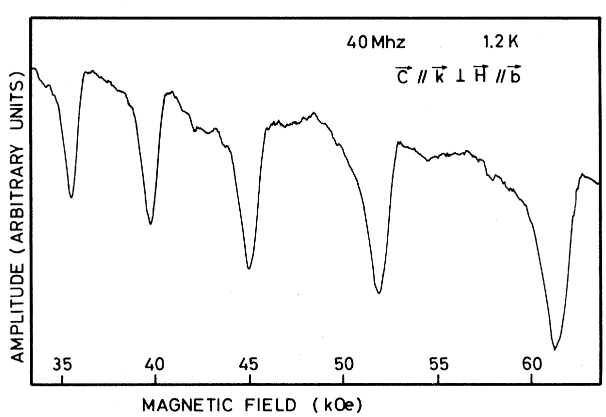

FIG. 1. Recorder tracing of the amplitude variation of the delayed rf pulse (inversely proportional to the Rayleigh-wave attenuation). The magnetic field is normal to the metal surface and the Rayleigh wave propagates along the $\vec{c}$ axis. The Rayleigh velocity is 2.4 $\times10^5$ cm/sec.

It is interesting to study the shape of the peaks in order to obtain information about the effect of the surface on the conduction electrons. Figure 2(a) shows the peak located at the highest magnetic field for different frequencies at 1.2 K. The peak height is about 1.7 dB/cm at 40 MHz, 2.7 dB/cm at 80 MHz, and 4 dB/cm at 120 MHz. The linewidth at half-height is 1500 ± 80 Oe at 40 MHz and 1830 ± 90 Oe at 80 MHz. So there is a broadening of the line as the Rayleigh-wave frequency is increased. At 120 MHz the signal-to-noise ratio is small and it is not possible to know from the recorder tracing if the line is broadened still further. This line broadening as the frequency is increased is unexpected. The temperature dependence of the linewidth has been also studied. There is no change in the line shape between 0.4 and 1.2 K. At 4.17 K and 40 MHz the peak height is about 1 dB/cm and the linewidth is 1800 ± 100 Oe. So, up to 4.17 K, the line shape is not very sensitive to the temperature variation.

Grishin and Kaner[1] have studied theoretically the shape of the absorption line in the case of specular reflections of the electrons on the metal surface. This hypothesis is justified because the GQO of the Rayleigh-wave attenuation are due to the electrons which have a small velocity projection on the magnetic field direction and for these electrons the scattering by a rough surface is close to specular. At zero temperature there are

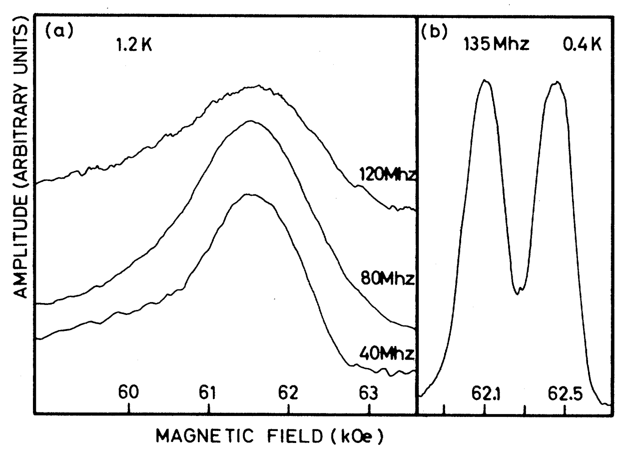

FIG. 2. (a) Shape of an attenuation peak for different frequencies. The magnetic field and the $\vec{b}$ axis are normal to the surface. The Rayleigh wave propagates along the $\vec{c}$ axis. The receiver gain is different for each curve. (b) Recorder tracing of the peak obtained by studying the GQO of the longitudinal bulk waves. The magnetic field and the acoustic wave vector are parallel to the $\vec{b}$ axis.

two limiting cases. The low-frequency case ($\omega\tau \ll 1$, where ω is the Rayleigh-wave frequency and τ the relaxation time of the electrons in the metal) leads to a linewidth

$$\delta H \sim H/(\chi l)^2, \qquad (1)$$

where χ is the damping decrement of the wave and l is the electronic mean free path. χ is about $0.4k$ in the isotropic case (k is the wave vector of the Rayleigh wave) and l is about 1 cm as it will be seen below. Equation (1) is valid only if the condition $N/(\chi l)^2 \ll 1$ is satisfied (N is the Landau-level number). Under the present experimental conditions $N=5$ for the peak located at 61.5 kOe and $(\chi l)^2$ is about 10^5 at 40 MHz. So we can use Eq. (1) which gives $\delta H/H \sim 10^{-5}$. This value is much smaller than the experimental one (2×10^{-2}). The high-frequency case ($\omega\tau \gg 1$) leads to a line which is still narrower than in the low-frequency case. Thus, it is not possible to explain the experimental linewidth with the theory at zero temperature. The effect of temperature on the line shape has also been considered by Grishin and Kaner.[1] It induces a line broadening which is similar to the thermal broadening in the case of bulk waves. Then the linewidth is[3]

$$\delta H = 3.53 kTHPm_c c/e\hbar, \qquad (2)$$

where P is the period in $1/H$ and m_c is the cyclotron mass. Equation (2) is valid only if the collision broadening is much smaller than the thermal broadening. Under the present experimental conditions $m_c = 0.6m_0$ and Eq. (2) gives $\delta H/H = 6\times10^{-3}$ at 1.2 K. This value is still smaller than the experimental one.

To conclude about any surface effect on the linewidth, it is important to make sure that there is no broadening effect in the bulk. Therefore I have also studied the GQO of the longitudinal bulk waves. It is easy to generate bulk waves with the comb used to generate surface waves. It is sufficient to put the comb out of tune (i.e., the frequency of the rf pulse is such that the corresponding Rayleigh wavelength is different from the tooth spacing). For certain frequencies, the comb generates bulk waves which are reflected on the opposite face and give rise to an acoustic echo on the same comb. Figure 2(b) shows the peak which has been obtained at 0.4 K with longitudinal waves. There is a spin splitting of the peak.[3] However the two subpeaks are well resolved and there is no appearance of collision broadening (which leads to an asymmetrical line shape). Hence we can conclude[7]

$$B \equiv \omega\tau(2kT/m_\parallel V_s^2)^{1/2} \gg 1, \qquad (3)$$

where $m_\parallel$ is the effective mass in the magnetic field direction and V_s is the sound velocity. Practically, the line has a symmetrical shape as soon as B reaches 3.[8] Taking this limit for B and using Eq. (3), we obtain $\tau > 0.4\times10^{-8}$ sec (I have taken $m_\parallel = m_0$ and $V_s = 4.1\times10^5$ cm/sec). Thus we are in the experimental case $\omega\tau \sim 1$ which is not surprising in very pure gallium. The width of the peaks for bulk waves varies as a function of temperature in agreement with Eq. (2) (taking the spin splitting into account). It is 630 Oe at 1.2 K and 1100 Oe at 4.17 K (for the peak located at 62.3 kOe). These results are to be compared with the linewidths obtained from Rayleigh waves. The temperature effect is quite different in the two cases. Furthermore the linewidth is always larger for Rayleigh waves than for bulk waves. The broadening of the peaks in the case of surface waves is probably due to diffuse reflection of the electrons on the metal surface. This hypothesis seems to be consistent with the too-small dependence of the linewidth with the temperature. It seems also consistent with the observed broadening of the line as the frequency is raised from 40 to 80 MHz. The elastic field penetrates indeed into the metal less deeply for an 80-MHz Rayleigh wave than for a 40-MHz one and so the attenuation of Rayleigh waves by the electrons may be more sensitive to defects and strains which are present near the surface.

The GQO of Rayleigh waves in transverse magnetic field seem to be promising for the study of surface effects in metal. The experimental results cannot be explained with the theory of Grishin and Kaner who have treated the problem in the case of specular reflections. Actually, the metal surface is not perfect and the magnetic field is not exactly normal to the surface (the deviation from the normal is less than 1° but the exact deviation is not known more precisely). This last point may be important in regard to the specular-reflection hypothesis. Further experiments are in progress to attempt to resolve the linewidth problem.

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5*f*-Electron Excitation Energies and the Coulomb Term, *U*, in the Light Actinide Metals

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Relativistic Hartree-Fock-Wigner-Seitz band calculations have been performed for the actinide metals Ac through Am in order to estimate 5*f* excitation energies. Our calculations predict that the tetravalent state (i.e., four *s-d* conduction electrons) is favored for the lighter elements with a crossover to a trivalent ground state occurring near uranium. We find the Coulomb energy, *U*, for 5*f* electron hopping to increase from 2–3 eV at Th to 4–5 eV at Am.

The 5*f* electrons of the actinide metals are of considerable interest for the study of magnetism since they are less localized than the 4*f* states of the rare earths but less itinerant than transition-metal 3*d* electrons. One quantity particularly relevant to magnetic behavior is the Coulomb term *U*, the energy required for an extra magnetic electron to hop onto an atomic site. In this Letter we report the first theoretical estimates of *U* for the light actinides, Ac through Am. *U* is found to increase across the series, consistent with the view[1] that the 5*f* electrons become more localized with increasing atomic number. Essential to the calculations are estimates of 5*f* excitation energies, which are interesting in light of recent x-ray photoemission studies[2] of thorium, uranium, and some of their oxides; in anticipation of further experimental progress we also report estimates of these 5*f* excitation energies. We employ a computational scheme[3] which has been quite successful in predicting 4*f* excitations in the rare-earth metals and, as experimental data become available, it will be interesting to assess the applicability of the method to the actinides.

Central to the present investigation is the neglect of hybridization between the 5*f* and the 6*d*-7*s* conduction electrons. The 5*f* states are degenerate with the conduction bands and hybridization is assuredly significant in these metals[1]; but our aim is to estimate the excitation energy of a single 5*f* electron as a function of 5*f* occupation number, and for this purpose it is essential to keep the 5*f*'s distinct from the conduction electrons by setting hybridization equal to zero. The atomic configuration $5f^n 6d^{m-1} 7s$ is assumed appropriate to the metal, and we perform calculations for varying 5*f* occupancy *n* (with a concomitant change in the valence *m* to preserve charge neutrality). *n* is confined to integral values so that atomic spectral data may be used to estimate correlation effects. Band calculations[4-6] are performed, and by permitting the 6*d* and 7*s* counts to be nonintegral, the bands and their associated charge densities are iterated to crude self-consistency.[6] The total Hartree-Fock band energy E_{band} and one-electron energies ϵ_i are evaluated. During the calculations the positions and widths of the *occupied* *f* levels are monitored. The level widths decrease with increasing atomic number; for example, they are 2 eV and 0.7 eV for trivalent Th and Am, respectively.