Dynamics of the iridium lattice: experiment and calculation

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(Submitted 26 November 1987; resubmitted 12 January 1988)
Pis'ma Zh. Eksp. Teor. Fiz. **47**, No. 5, 245–248 (10 March 1988)

A pseudopotential model is used to describe the static and dynamic characteristics of the iridium lattice. The phonon state density in iridium has been determined experimentally for the first time. The results are compared with calculations.

1. Iridium is a typical transition metal with a high density of d states at the Fermi level. This circumstance is the primary difficulty in the theoretical description of the lattice properties of iridium. So far, only two calculations of the Ir phonon spectra have been carried out.[1,2] Mehrotra *et al.*[1] used a model expression for the dynamic matrix to calculate the phonon dispersion $\omega(\mathbf{q})$ and the spectral density $F(\omega)$. Tréglia and Desjonquéres[2] used a slightly different model and a recursion method to determine $F(\omega)$ (only). The phonon spectra found in Refs. 1 and 2 differ substantially in shape and in the limiting frequency $\omega_{\max}$.

There have been no experimental studies of the dynamics of the Ir lattice. This void hinders the theoretical work on the lattice dynamics as well as on the electron-phonon interaction and the kinetic properties of transition metals. We have accordingly undertaken a theoretical and experimental study of several lattice properties of Ir.

2. In the measurements of $F(\omega)$ we used a cold-neutron source and a multidetector time-of-flight spectrometer for a polycrystalline sample. Figure 1 shows the Ir phonon spectrum reconstructed from the doubly differential cross section for inelastic scattering of neutrons. Multiple and multiphonon processes have been taken into account. The low-frequency part of the spectrum was reconstructed from data on the low-temperature heat capacity.[3] This spectrum has a well-defined two-peak structure with maxima of equal intensity at frequencies of[2)] 6.4×10^{-4} a.u. and 8.5×10^{-4} a.u. and a fairly sharp spectral boundary at 10.5×10^{-4} a.u.

3. The theoretical model which we used to calculate $F(\omega)$ is described in Ref. 4. It is based on a representation of the binding energy of metals with s and d valence electrons in a form which uses a perturbation theory in the pseudopotential for s electrons. The contribution from the interaction of d electrons was modeled by a short-range Born-Mayer interatomic interaction potential. The ion charge Z was found as the number of valence electrons per atom which could be regarded as nearly free. The quantity Z was accordingly taken to be the sum of the effective occupation number of the s states (0.46) and of the charge outside the MT sphere (1.50), both found from band-theory calculations.[5] In the present study we used the Animalu-Heine-Abarenkov local model pseudopotential. Exchange-correlation effects were incorporated in the Taylor approximation. This model has four adjustable parameters, which were found through a least-squares fit of the experimental values of $\omega_{\max}$ and the elastic moduli B_{ik} at the equilibrium volume.

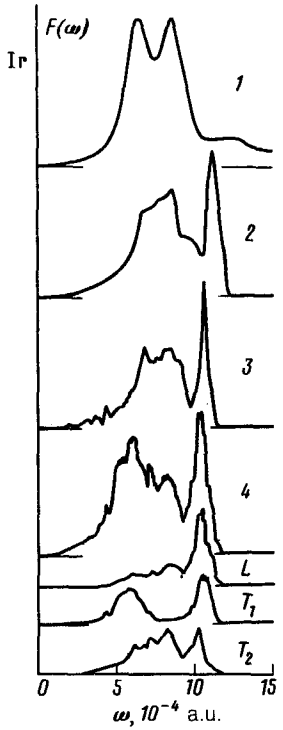

FIG. 1. Complete (1–4) and partial (L,T_1,T_2) phonon state densities in Ir. 1—Present experiments; 2,3—calculations of Refs. 2 and 1, respectively; 4—present calculations of L, T_1, and T_2.

The calculated static characteristics agree well with the experimental data. The discrepancies in terms of the values of B_{ik} do not exceed 15%. The calculated P-V diagram agrees with the experimental diagram, within the experimental errors, up to 600 GPa (Fig. 2). This agreement is consistent with the assumption that the interaction of the core states in Ir is negligible over a broad pressure range, and the renormalization of the parameters of the model by the pressure is unimportant. In the present

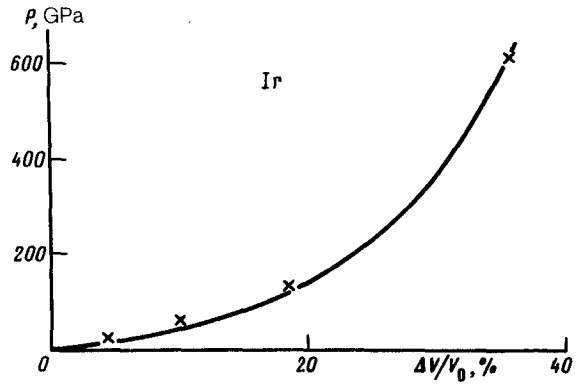

FIG. 2. P-V diagram of iridium. Solid line—Present calculations; crosses—experimental data.[6]

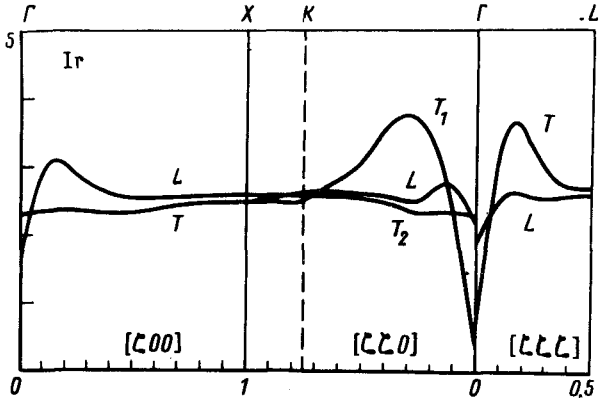

FIG. 3. Microscopic Grüneisen parameters of iridium.

study we calculated the barik derivatives of the elastic moduli: $dB_{11}/dP = 6.50$, $dB_{44}/dP = 3.54$, and $dB_{33}/dP = 0.39$. Analysis of the experimental P-V diagram at low P yields[7] $dB_{11}/dP = 6.0$. This figure agrees with the value which we found. No experimental information is available on the pressure dependence of the shear moduli.

The calculations of $\omega(\mathbf{q})$ yielded smooth curves. The only anomalous feature was a negative dispersion at small wave vectors for the T_1 $[\xi\xi 0]$ branch. The same anomalous feature is observed experimentally for two fcc metals which are related to iridium: Au and Pt. Near the faces of the Brillouin zone, our calculated curves are 10–15% lower than the results of Ref. 1. These results are in better agreement with the experimental results, although the longitudinal branch in our calculations is about 10% too high near the face of the Brillouin zone. Spectra 1–4 in Fig. 1 have values $\omega_{\max} = (10.5,\ 12.2,\ 11.7,\ \text{and}\ 11.5) \times 10^{-4}$ a.u. and $\langle\omega\rangle = (7.45,\ 8.54,\ 8.27,\ \text{and}\ 7.32) \times 10^{-4}$ a.u., respectively. Consequently, although all the calculations overestimate $\omega_{\max}$, the model which we have used gives a better description of the iridium phonon spectrum than that given by the models of Refs. 1 and 2.

4. We have calculated the microscopic Grüneisen parameters, $\gamma(\mathbf{q}) = -\delta \ln \omega(\mathbf{q})/\delta \ln V$, in Ir for the first time (Fig. 3). A corresponding calculation has been carried out previously for copper.[8] The calculations show that the pressure dependence of the frequencies in Ir is considerably stronger than that in Cu, although the $\omega(\mathbf{q})$ spectra themselves are nearly identical for these two metals. Furthermore, the relation $\gamma_T \gtrsim \gamma_L$ holds at most points in the Brillouin zone of Ir, while in Cu we have, on the contrary, $\gamma_T < \gamma_L$. Data from tunneling spectroscopy yield $\gamma_T > \gamma_L$ for essentially all of the entities studied in Ref. 9 (for Ir there are no experimental data on the pressure dependence of the tunneling spectra). On the whole, the anisotropy of the differential characteristics of the Ir phonon spectrum is more pronounced than that of Cu.

In summary, the experimental data on the statics and dynamics of the Ir lattice can be described satisfactorily in the pseudopotential model of the binding energy[4] with a d-electron interaction.

We wish to thank V. G. Vaks for many useful discussions and A. V. Khotkevich for consultation and for practical assistance.

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[2)]1 a.u. = 2 Ryd = 27.2 eV.

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Translated by Dave Parsons