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OSCILLATORY STRUCTURE IN MEASURED TOTAL $Li^+ + Li$ CHARGE TRANSFER AND COMPARISON WITH THEORY*

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Measured $Li^+ + Li$ total charge-transfer cross sections are reported and compared with the computed results of Peek, Green, Perel, and Michels based on an ab initio two-state calculation. Both experiment and theory show oscillatory structure in the cross sections with very good agreement in cross-section magnitude and oscillatory structure. There exist, however, small but important differences in the oscillation characteristics.

An ab initio calculation was made on the two lowest potential states of Li_2^+ as a function of the internuclear separation by Michels.[1] The difference between these two potential curves was used as a basis for a calculation of the total resonant lithium charge-transfer cross section.[2] With use of a two-state expansion within the framework of the impact-parameter method, the calculated cross section as a function of the velocity shows regular, large-amplitude (~10%) oscillations as a result of the potential-difference curve containing a maximum at a large impact parameter.[3] The oscillations are similar to those seen in theoretical[4] and experimental[5] investigations of cesium resonance. The computed lithium cross-section values and oscillation characteristics showed good correlation with preliminary measurements. In this paper the final measurements are reported and the results compared with the calculations.

The measurements were made using a crossed-beam technique[5] with data taking facilitated by an XY plotting method[6] and with ion generation and atom detection accomplished using surface ionization by an oxygenated surface.

The ion source[7] consisted of a reservoir of lithium which was heated to above the melting point with lithium transported by capillary forces to a vaporizer region. The vaporizer region temperature was used to control the lithium feed rate to a porous tungsten ionizer which was heated to about 1100°C. A directed oxygen spray was used to increase the surface work function in order to enhance the ionization efficiency of the source. Usable ion beam currents ranged from 0.1 to 1 μA during cross-section measurements.

Atomic-beam intensity measurements depend upon surface ionization to efficiently convert the atomic flux into an equivalent ion current using an indirectly heated tungsten surface. The ionization efficiency of the atom detector has a direct effect upon the accuracy of the cross-section measurements, therefore detection efficiency was carefully examined. Desorption time measurements were made as a function of the surface temperature and degree of oxygenation to help evaluate the accuracy of the ac method used

to detect the modulated atomic beam.[8]

Uncertainty in the desorption was estimated to contribute an error of no more than ±5%. Ionization efficiency was examined by also varying temperature and oxygenation. For different degrees of oxygenation, the detection current increased with temperature, then asymptotically approached a maximum value. With increasing oxygen (at a fixed temperature), the current increased, then decreased when the chamber pressure was sufficiently high to attenuate the atomic beam ($\sim 10^{-5}$ Torr). From these considerations, the error in determining the atom intensity was taken to be less than ±15%. Since this was the largest uncertainty in the measurement, the accuracy of the cross-section magnitude was estimated at ±15%.

The oxygen flow required for efficient Li surface ionization increased the vacuum-chamber pressure somewhat above the operating level for previous cross-section measurements. The pressure and the high source temperatures caused more noise than usual. At low signal levels (which occur at low ion-accelerating voltages) the noise introduces nonlinear effects, particularly at the log multiplier used to obtain the ratio of the charge-transfer signal to the ion-beam current. Similar effects occur at high voltages from ion-source arcing due to the oxygen pressure. The overall effects from these two can distort the cross-section curve shape, particularly at the high and low ends of the ion velocity range.

Figure 1 shows the measured $Li^+ + Li$ cross section as a function of the ion velocity (and energy) along with the experimental results of Lorents, Black, and Heinz.[9] The two cross-section curves agree within experimental error and exhibit the typical resonant decrease with increasing velocity. The superimposed oscillations are similar to those observed with the other resonant cross sections.[5,10,11]

For a clear display of the oscillatory structure, the cross section is shown in Fig. 2 as a function of inverse velocity, along with the calculated cross section of Peek, Green, Perel, and Michels.[2] The agreement in cross-section magnitudes is very good. The experimental curve shows an increase at the two ends of the measured range which is probably due to the curve distortion uncertainty discussed above. This cross section deviates somewhat from the curve shape $\sigma^{1/2} = A - B \ln v$ obtained with all the rest of the alkali-metal resonance cross sections measured in this laboratory. It is estimated that the uncertainty in the relative cross section and curve shape below 0.6 keV (>17 a.u.) is about 15% so that the theoretical results are within the uncertainty of the measurement.

There are, however, differences in the oscillatory structures of the two curves which are examined using the stationary-phase approximation

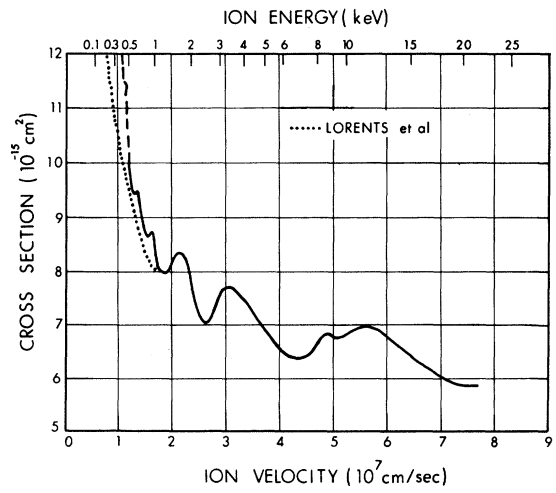

FIG. 1. Measured lithium resonance charge-transfer cross section as a function of the ion velocity and energy, showing clear oscillatory structure. The experimental results of Ref. 9 are shown to be in good agreement with our results except that they did not observe oscillations. The dashed portion of our cross-section curve has a greater uncertainty than the rest of the curve.

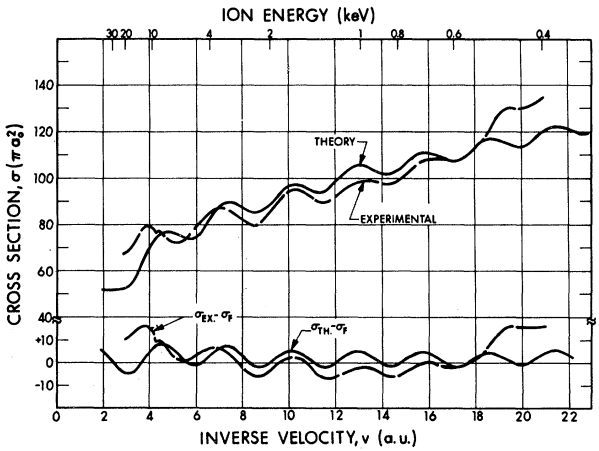

FIG. 2. The measured cross section is compared with the theoretical results of Ref. 2, on a plot of the cross section versus inverse velocity. The uniformity of the oscillations and the similarity between the two curves are clearly displayed. The insert shows the oscillatory parts obtained by subtracting the smoothly varying theoretical part (σ_F) from both cross sections.

of Smith.[4] He showed that the cross section could be characterized by a smoothly varying part of an oscillatory part. The oscillatory part of the cross section can be approximated by

$$-\alpha(v)\cos\pi(\beta v^{-1}-\delta), \quad (1)$$

where $\alpha(v)$ is the amplitude, β is the frequency, and δ is the phase constant of the oscillations. Note that this is a negative cosine curve so that maxima and minima occur for odd and even integer values of n, respectively, where $n=\beta v^{-1}-\delta$.

Oscillation frequencies and phase constants were obtained from Fig. 3 which is a plot of n vs v^{-1}. Maxima and minima occur at odd and even values of n, on the assumption that the oscillatory part of the cross section is approximated by the damped cosine curve given by Eq. (1). The theoretical data lie on a straight line with $\beta=0.71$ a.u. (1.55×10^8 cm/sec) and $\delta=0.2$. Considering that the velocity at the experimental datum point $n=13$ is the most uncertain, the slope (β) above $v=10$ a.u. is the same as the theoretical line with $\delta=0.4$. Below 10 a.u., the slope is lower with $\beta=0.61$ (1.33×10^8 cm/sec) and $\delta=0.7$. The phase constants (δ) differ from those of the rest of the alkali-metal resonance measurements which yield approximately $+\frac{3}{4}$. For high-velocity collisions, such as those for Li above a few keV ($v^{-1}\sim10$ a.u.), Smith[12] suggests that a phase change can occur. This may explain the change in the experimental slope in Fig. 3. It should be noted that the calculations do not include the high-velocity effects which appear to be important below 10 a.u.

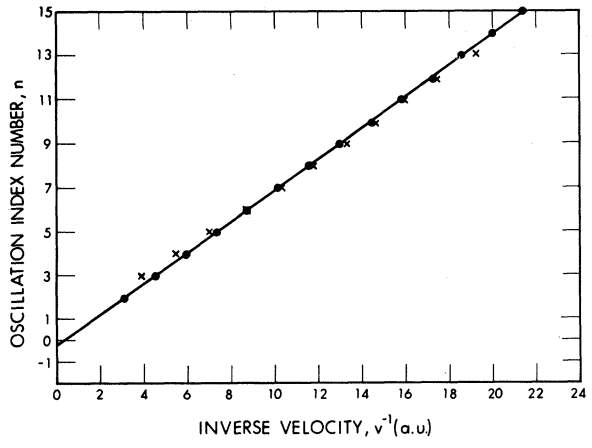

FIG. 3. Oscillation maxima and minima plotted as n vs v^{-1}. The theoretical data (solid circles) lie on a straight line. The experimental data (crosses) have a lower slope at lower inverse velocities and show very good agreement with theoretical data at higher inverse velocities. These plots were used to determine values of β and δ.

The theoretical oscillation amplitude is approximately given by $\alpha(v)=12.5v^{1/2}$ (in a.u.). This shows the same velocity dependence obtained by Smith[4] and Marino[13] in calculations for Cs resonance calculations. As in the Cs case, the Li experimental amplitudes fall off more rapidly than theoretical. This may result from damping in the probability oscillations not accounted for in the calculations.

The present Li resonance charge-transfer measurements show cross-section values and oscillation amplitudes in good agreement with computations based upon ab initio interaction potentials. The experimental oscillation characteristics differ from those of the calculations below $v^{-1}\sim10$ a.u. The use of a two-state impact parameter calculation does account for most of the features of the Li resonance cross section. It should be pointed out that the agreement between theory and experiment verifies the ab initio and cross-section calculations. However, a unique potential-difference function cannot be determined from a total cross-section measurement because rather different functions can provide the same oscillation characteristics.[12]

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HYPERFINE ANOMALY IN ^{193}Ir BY MÖSSBAUER EFFECT, AND ITS APPLICATION TO DETERMINATION OF THE ORBITAL PART OF HYPERFINE FIELDS*

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A 7% hyperfine anomaly is found in Mössbauer measurements of the 73-keV transition in ^{193}Ir. A 2% difference is found between the anomalies in antiferromagnetic IrF_6 and Ir-Fe alloy. The difference is ascribed to orbital contributions which differ for the two iridium environments. By using known details of IrF_6 we find the orbital field in the Ir-Fe to be $H_l = +335 \pm 200$ kOe.

We have observed a 7% hyperfine anomaly between the ground state and first excited state in ^{193}Ir by Mössbauer effect measurement, and a difference between the anomalies for iridium in ferromagnetic and antiferromagnetic environments. We are able to use the results to partition the hyperfine field at ^{193}Ir in an Ir-Fe alloy into a contribution due to a Fermi contact interaction and one due to noncontact fields, the latter being primarily orbital in origin. It is chiefly upon this new magnetic technique that we wish to report. The measurements of the anomaly will be treated briefly here and in detail in a later publication.[1]

Anomalous hyperfine interaction describes the difference between a nuclear magnetic moment measured in a uniform field and the moment measured in a magnetic field of hyperfine origin.[2,3] Since a hyperfine field is observable only in a product with a nuclear moment, the anomaly is observed by comparing the ratio of two moments or nuclear g factors measured in the two types of field. Many such anomalies between the ground states of a pair of isotopes have been measured, for example by atomic-beam techniques. Grodzins and Blum[4] made the first Mössbauer type measurement of the anomaly between two states of the same nucleus (the ground state and first excited state in ^{57}Fe), where it was found to be marginally small.

A large anomaly might be expected to be present in the iridium nuclei ^{191}Ir and ^{193}Ir.[5] The relatively large size of the nucleus results in a considerable variation of electron density over the nuclear radius. The contact hyperfine field, which depends on the density, is thus variable over the radius. The orbital and spin motions of the unpaired nucleons cause different radial distributions of magnetic moment and therefore contribute differently to the overall interaction in the radially varying density. The $\frac{3}{2}^+$ ground state of the odd-proton nucleus ^{193}Ir, for example, is favored for the effect because insofar as it is $d_{3/2}$ in character, the orbital and spin contributions to the nuclear moment tend to cancel, and a minor difference in the separate interactions shows up as a relatively large effect. The $\frac{1}{2}^+$ excited state at 73 keV is not so especially favored. It is the transition between these two states which we use.

We measured the ratio g^*/g^0 of excited-state to ground-state nuclear g factors in an external field of 73 kG supplied by a superconducting magnet. The absorber was iridium metal and the source ^{193}Os from neutron capture in ^{192}Os metal. The source itself has a small quadrupole splitting[6] which was taken into account in the fitting. The spectrum is incompletely resolved and contains two prominent outer lines and two weak inner ones. The separation of the prominent lines gives $g^* + g^0$. For g^0 we can take Narath's value[7] uncorrected for Knight shift and diamagnetism, namely $g^0 = +0.10589$. This is permissible since both measurements are done in external fields and on the same substance. With this we obtained $R_u \equiv g_u^*/g_u^0 = +9.51 \pm 0.03$. The subscript u indicates measurement with a field that is uniform over the nuclear volume.[8] This is to be compared with the same ratio measured in a 2.7-at.% alloy of Ir in Fe by Wagner et al.,[9] who obtained $R_{Fe} = +8.875 \pm 0.018$. As the measure of the anomaly we take

$$\Delta_{Fe} \equiv (R_u - R_{Fe})/R_{Fe} = 0.072 \pm 0.004. \quad (1)$$