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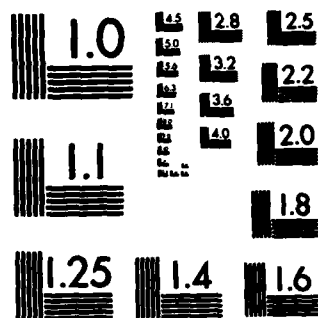
LOW-ENERGY COLLISIONS EXCITED ATOMS(U) LA JOLLA INST CA 171
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A summary of research performed under AFOSR Contract No. F49620-82-K-0023 is given. The contract covered the period 1 May 1982-30 April 1984. The report describes molecular-beam studies of ion-pair production, charge transfer, and measurements of the fraction of excited Na atoms in a composite beam of ground-state and excited Na atoms. Some of the experiments involved laser excited Na as a reactant. Included are investigations of the Na-Br, doubly charged Ar-Ar, Ne ion-metastable He, excited Na-Na, and Li-Na systems.

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PROGRAM

Selected ion-pair production, chemi-ionization, and resonant and near-resonant charge-transfer reactions involving excited atoms will be experimentally studied in a range of relative energy from thermal, or threshold, to several hundred electron volts. Reactants of the processes include metastable rare-gas atoms, rare-gas ions, halogen atoms, ground-state and excited alkali atoms, and alkali ions. Reactions leading to Li^- are of special interest. The studies will be conducted in a merging-beams apparatus. A laser system will be used in conjunction with this apparatus for experiments involving some excited atoms such as $\text{Na}(3^2\text{P}_{3/2})$. Cross sections as a function of collision energy, threshold behavior where applicable, and product-energy distributions will be measured. Existing theories, such as the Landau-Zener-Stueckelberg curve-crossing model for ion-pair production and the Demkov approach for near-resonant charge transfer, will be used to explain the observed data. Attempts will be made to modify these theories to account for discrepancies, new theories will be discussed where possible, and the need for additional theoretical effort will be noted.

ACCOMPLISHMENTS

The research accomplished on the Contract F49620-82-K-0023 for the period 1 May 1982 - 30 April 1984 is cited below.

1. Absolute and relative cross sections were obtained for the ion-pair production process $\text{Na} + \text{Br} \rightarrow \text{Na}^+ + \text{Br}^-$ in which the reactants and products are in the ground state. The studies were made by a merging-beams technique in a range of relative kinetic energy W of the reactants from the threshold of 1.78 eV to 500 eV. Agreement is excellent between the experimental results and calculations of Faist and Levine.

The purpose of conducting this experiment was three-fold. First, the basic physics of this ion-pair production process is interesting and particularly the threshold behavior, which is different from that of other

alkali-halogen systems previously measured. Second, the experiment would give us experience in studying this type of process. Such experience would be useful in investigating more complicated processes of ion-pair production involving excited and ground-state (g. s.) alkali atoms of special interest to the Air Force. Finally, the reaction could be used in measuring the fraction of excited Na in composite beams of excited and g. s. Na atoms. Later we see that for small fractions (i. e. , < 30%) another reaction was more suited to this purpose.

Our results of this study have been submitted for publication.

2. A surface-ionization Li^+ source has been developed. A Li^+ beam from this source has been neutralized in a Na vapor cell resulting in a Li neutral beam. It was intended that such a beam be used for studies of Li interacting with an excited Na beam.

3. A rather small fraction of our time has been devoted to the study of the reaction $\text{Ar}^{2+} + \text{Ar} \rightarrow \text{Ar} + \text{Ar}^{2+}$, a resonant charge-transfer process with a doubly ionized atom. This experiment is a demonstration of our continuing interest in charge-transfer reactions. In fact, the Na-Br ion-pair production reaction mentioned above can also be considered to be a charge-transfer process.

The results of the Ar^{2+} -Ar study are in reasonable agreement with the predictions of Fetisov and Firsov and have been published in Chemical Physics Letters. The measurements were made for $2 \leq W \leq 1000$ eV.

4. An investigation was conducted on the reaction $\text{Ne}^+ + \text{He} (2^3\text{S}) \rightarrow \text{Ne}^* + \text{He}^+$, where Ne^* represents excited Ne. The measurements were made for $0.01 \leq W \leq 1000$ eV. The results are in reasonable agreement with calculations obtained from a modified Demkov approach. This study adds to the rather small bank of data on charge-transfer reactions at low energies involving excited atoms. The results have been published in Chemical Physics Letters.

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5. Our goal during the past year has been to prepare for studying the reaction $\text{Li} + \text{Na}^* \rightarrow \text{Li}^- + \text{Na}^+$, where Na^* represents excited Na in the $3p\ ^2P_{3/2}$ state. This reaction could eventually result in the production of intense Li^- beams and finally, through stripping, to equally intense Li neutral beams. The latter are of importance in Air Force applications.

One of the first requirements in achieving this goal was to produce a fast (several keV) Na^* beam. This was done by exciting a fast beam of g. s. ($3s\ ^2S_{1/2}$) Na atoms with a laser. The g. s. atom beam was produced by charge transferring Na^+ from a surface ionization source in a vapor of Na atoms. A single-frequency CW dye laser pumped by an Ar ion laser was used for the excitation. The laser was tuned to the Doppler shifted D_2 line ($5890\ \text{\AA}$) of Na. In fact, it was tuned for the hyperfine transition from $M_F = 2$ in the g. s. to $M_F = 3$ in the excited state.

An advantage of exciting a fast atom beam with a laser is that the Doppler width is considerably reduced from that obtained by exciting a thermal beam. In fact, for our 1300 K source temperature (corresponds to 0.11 eV) and an atomic beam energy of 5000 eV, a reduction of 2.4×10^{-3} spread in the velocity of the beam due to the source is calculated. The resultant spread due to the source $\Delta v_g = 230\ \text{cm/s}$, which is equivalent to a frequency change of $\Delta \nu_g / \lambda = 230 \div (5890 \times 10^{-8}) = 3.9\ \text{MHz}$. This Doppler width is well within the 10 MHz natural linewidth of the Na- D_2 line as is the 1 MHz linewidth of the laser beam. Therefore, the laser was capable of exciting all of the g. s. $M_F = 2$ atoms in the beam and, in fact, saturating the beam since calculations indicate each atom would be excited 60 times during its passage through the interaction region. On this basis, it was calculated from the statistical weights of the hyperfine levels that $5/8 \div 2 = 5/16$ or 31% of the atoms in the interaction region should be in the upper level.

An experiment was devised to measure $\Delta \nu$, the linewidth of the atomic beam. This width is a composite of $\Delta \nu_g$, a spread due to angular

divergence Δv_a , a spread due to power broadening Δv_p , and the natural linewidth of the Na-D₂ line Δv_n . At 5000 eV, the Δv is 55 MHz, which reduces to a spread of 37 MHz for the combined effects of Δv_s and Δv_a (after Δv_p and Δv_n were taken into account). The angular divergence of the experiment, or half angular spread, is about 5 milliradians, and computations indicate that very little of the 37 MHz is contributed by this spread. In fact, these calculations show that 36 MHz is attributable to the source (i.e., velocity spread of atoms from the source).

The significance of the above figures is the following. First, the measured linewidth of 55 MHz is quite large, and instead of exciting 31% of the atoms in the beam, the laser will excite only $(\Delta v_n / \Delta v) 31 = (10/55) 31 = 6\%$. This can be improved only by decreasing the velocity spread of atoms from the source since the other spreads are near their realizable minimums. It is not clear why the surface ionization source produces a spread of 36 MHz instead of its predicted 3.9 MHz, but presumably the trouble originates from resistivity in the fused, silica glass from which Na⁺ emerges and a variable work function of the glass. Our future plans are to design a surface ionization source that does not use such glass. Rather, we plan to let Na vapor impinge and surface ionize on a hot, porous W plug. This laboratory has used such a source in the past.

While designing and developing the source, we decided to conduct experiments with the fused glass source and 6% excitation. These would not be beam-beam experiments but rather, because of the small percentage of excited atoms, beam-gas efforts. Elastic scattering is a bigger problem in beam-gas than in beam-beam experiments, but can be mollified by using larger collision energies, say in the range of several hundred to several thousand electron volts. This presents no problem for the Air Force study of producing negative ion beams since such beams have to be generated at fairly large energies in order to achieve sufficient intensity. Before such experiments could be done

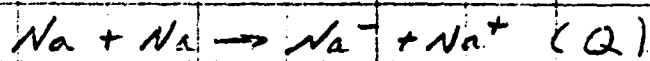
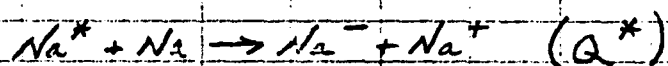
completely quantitatively, the actual fraction of excited atoms had to be measured. The next section describes how we accomplished that goal.

6. Actual measurements of f^* , the fraction of atoms in a beam excited by a laser, are rather rare in the literature. Generally, it is assumed that the laser saturates the excitation, and a calculated value of f^* is used. If a measurement is made, it is usually of the intensity of the fluorescence associated with the excitation. Rather than trying to measure photon intensities to determine f^* for our Na beam, we felt we could get a more accurate value through the use of a chemical reaction. We have used a similar technique in the past to determine the fraction of metastable atoms in a composite beam of excited and g. s. rare gas atoms. The trick is to find a reaction which proceeds with g. s. atoms but not with the excited atoms whose fraction is being measured. The f^* is then determined by measuring reaction products with the source of excitation on and off - in the present case, the laser. In our case of a composite beam of g. s. Na and Na^* , we concentrated on ion-pair producing beam-gas reactions, i. e., those reactions which produce a positive and a negative ion. (We had originally planned to use the beam-beam reaction $\text{Na} + \text{Br} \rightarrow \text{Na}^+ + \text{Br}^-$ for measuring f^* , but only 6% excited atoms obviated this approach.) Such considerations require that the sodium beam react with a gas which has a relatively large electron affinity (EA). Gases which were tried include O_2 , NO_2 , Br_2 , and I_2 . We settled on I_2 , which has an EA = 1.72 eV. Ion pairs of Na^+ and I_2^- are formed when the reactant covalent potential curve of g. s. Na and I_2 crosses the product ionic curve of Na^+ and I_2^- . The covalent curve of Na^* and I_2 crosses the Na^+ and I_2^- curve at such a large internuclear distance (and hence with negligible coupling) that no interaction and, hence, no I_2^- occurs. The f^* measured with this reaction indeed agrees with the calculated value of about 6%. We plan to publish a paper on this technique. The first experiment we conducted using a Na^* beam is briefly described next.

7. We decided to investigate the ion-pair producing reaction $\text{Na}^* + \text{Na} \rightarrow \text{Na}^- + \text{Na}^+$ before the Li reaction because we had a cell for producing Na vapor and not one for Li. The vapor in such a cell is the gas that is reacted with the fast Na^* beam. Not only did we measure absolute and relative cross reactions Q^* for this process but also for $\text{Na} + \text{Na} \rightarrow \text{Na}^- + \text{Na}^+$, where all species are in the g. s. Figure 1 shows the results of some of these experiments. The graph shows Q^*/Q versus W , where Q^* and Q are the cross sections for ion-pair production for collisions of Na^*-Na and $\text{Na}-\text{Na}$, respectively. The results were obtained by measuring Na^- generated from the fast beam of atoms. It is clear from the figure that ion-pair production is greatly enhanced by exciting the Na, and we anticipate that the same will be true in the case of $\text{Li}-\text{Na}^*$ collisions.

One other aspect of the Na^*-Na ion-pair producing system that is interesting and that we plan to study is the competition between a single and double electron rearrangement in the formation of Na^- . That is, the reaction of Na^* and Na can be expressed as $\text{Na}^* + \text{Na} \rightarrow \text{Na}^- + \text{Na}^+$ and $\text{Na}^* + \text{Na} \rightarrow \text{Na}^+ + \text{Na}^-$. In other words, the Na^- product, i. e., $\text{Na}^-(3s^2\ ^1S_0)$, can originate either from the reactant Na^* or Na. If it comes from Na, an electron attaches to the 3s shell, and this is a one electron process. If it comes from Na^* , not only must an electron attach itself to the 3s shell but the 3p electron of Na^* must go down into the 3s shell resulting in a two electron rearrangement. The two electron scheme is presumably less likely. Initial measurements indicate that this is true.

8. Another preparation we made for the $\text{Li}-\text{Na}^*$ experiment was a study of the reaction $\text{Li} + \text{Na} \rightarrow \text{Li}^- + \text{Na}^+$, where all the species are in the g. s. Knowledge of this process is necessary because in the $\text{Li}-\text{Na}^*$ study only a fraction of the Na beam will be excited. Most of it will be in the g. s., and the contribution to Li^- from the g. s. atoms must be known. The EA of Li and ionization potential of Na are such that the crossing radius of the covalent Na-Li and ionic Na^+-Li^- curves is 3.2 Å. This is



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$Q^*/2$

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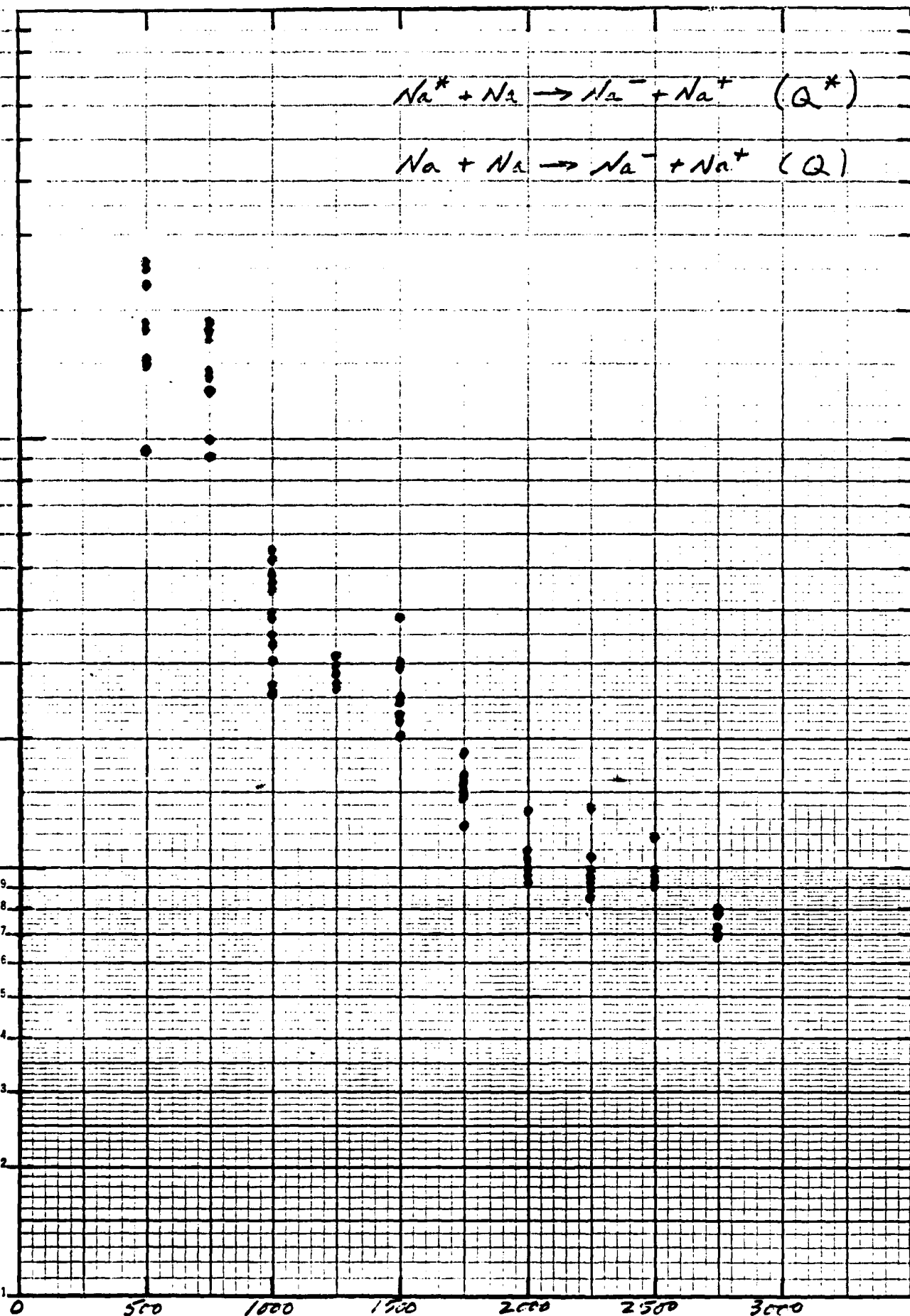


Fig. 1

Relative Energy, W (eV)

relatively small and leads to a large coupling potential making it difficult to transfer ultimately from the covalent to the ionic curve. Thus, a small Q for ion-pair production is expected. Figure 2 is a plot of Q versus W , and indeed verifies the expectation. The results were obtained by measuring Li^- produced when a fast Li beam collides with a vapor of Na.

PUBLICATIONS

1. R. H. Neynaber and S. Y. Tang, "Symmetric-Resonance Charge Transfer of Ar^{2+} in Ar," Chem. Phys. Lett. 92, 556 (1982).
2. R. H. Neynaber and S. Y. Tang, "Charge Transfer Between Neon Ions and Metastable Helium," Chem. Phys. Lett. 100, 316 (1983).
3. R. H. Neynaber and S. T. Tang, "Ion-Pair Production in Collisions of Na and Br," submitted to J. Phys. B.

TALKS

1. R. H. Neynaber and S. Y. Tang, "Charge-Transfer Collisions of Ne^+ and Metastable Helium," XIII International Conference on the Physics of Electronic and Atomic Collisions, Berlin, p. 486, 27 July-2 Aug., 1983.

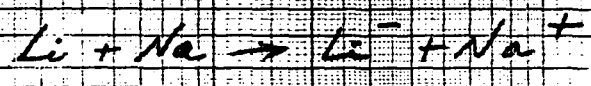
PARTICIPANTS

The participants in the research described above are Dr. R. H. Neynaber, Dr. S. Y. Tang, and Mr. D. P. Wang (graduate student).

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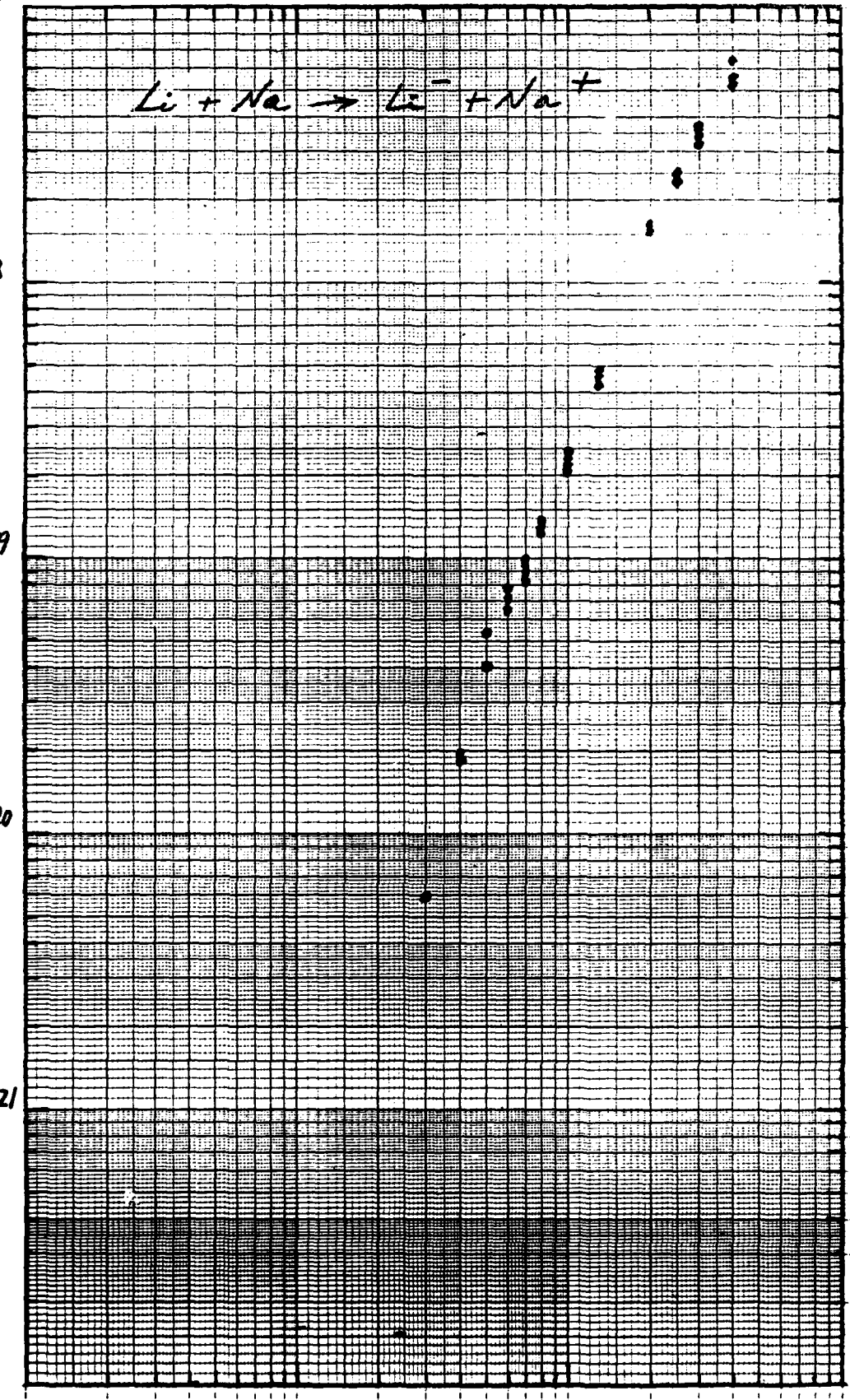


Fig. 2

Relative Energy, W (eV)

USE OF RESULTS

The Air Force Weapons Laboratory at Kirtland Air Force Base is interested in the production of Li beams and, thus, in any results associated with our forthcoming Li-Na^{*} study. Col. R. Zazworsky of Advanced Concepts/NTYP is especially close to this problem and the person at AFWL with whom we communicate. Also of interest is Dr. Lawrence Wright of Mission Research Corporation in Albuquerque. Dr. Wright is a theoretical physicist who studies ion-pair production and has a close working relationship with the Advanced Concepts Group at AFWL.

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