

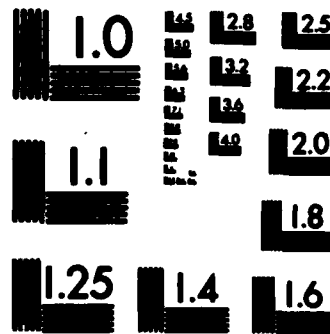
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19. ABSTRACT (Continue on reverse if necessary and identify by block number) <p>A summary of research performed under AFOSR Grant No. 83-0242 is given. The report is for the period 25 June 1983 - 24 June 1984. The review describes molecular-beam studies of ion-pair production and measurements of the fraction of excited Na atoms, <math>f^*</math>, in a composite beam of ground-state and excited Na atoms. The actual ion-pair production reactions studied are the following: <math>Na + Br \rightarrow Na^+ + Br^-</math>, <math>Na + I_2 \rightarrow Na^+ + I_2^-</math>, <math>Na(Na^*) + Na \rightarrow Na^+ + Na^-</math>, and <math>Li + Na \rightarrow Li^- + Na^+</math>. The reaction <math>Na + I_2 \rightarrow Na^+ + I_2^-</math> was used to measure <math>f^*</math>. A value of about 6% was obtained. The work was performed using instrumentation provided by the grant.</p>		19. ABSTRACT (Continue on reverse if necessary and identify by block number)	
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MATTHEW J. KERPER

Research Objectives Chief, Technical Information Division

Instrumentation purchased under the subject grant will be used to conduct particle-beam research of interest to the Air Force. Molecular beam techniques will be employed to study two-body reactions that will result in an intense, collimated beam of Li atoms at an energy of several hundred keV. Generating such a beam by charge transfer of a Li<sup>+</sup> beam is not efficient at this energy. It appears that it can most efficiently be produced by forming a Li<sup>-</sup> beam and stripping away valence electrons in a suitable gas. We plan to study reactions associated with the production of a Li<sup>-</sup> beam which is formed when a Li<sup>+</sup> beam interacts with a vapor of ground and excited (resonance state) Na atoms. One of the reactions (after Li<sup>+</sup> has been converted to Li) is  $Li + Na^* \rightarrow Li^- + Na^+$ . This will be examined by merging a Li beam with a beam of Na atoms excited with a single frequency dye laser. The instrumentation required to complete this study consists of some laser and optical components, some vacuum equipment, and a laboratory computer system for data acquisition and analysis. This apparatus will supplement as well as rejuvenate an existing merging-beam apparatus.

Status of the Research

The research accomplished on Grant AFOSR 83-0242 for the period 25 June 1983 - 24 June 1984 is cited below.

1. Absolute and relative cross sections were obtained for the ion-pair production process  $Na + Br \rightarrow Na^+ + Br^-$  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



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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  $\text{Li}^+$  source has been developed. A  $\text{Li}^+$  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. Our goal during the past year has been to prepare for studying the reaction  $\text{Li} + \text{Na}^* \rightarrow \text{Li}^- + \text{Na}^+$ , where  $\text{Na}^*$  represents excited Na in the  $3p^2P_{3/2}$  state. This reaction could eventually result in the production of intense  $\text{Li}^-$  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)  $\text{Na}^*$  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  $\text{Na}^+$  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 Å) of Na. In fact, it was tuned for the hyperfine transition from  $F = 2$  in the g. s. to  $F = 3$  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

$2.4 \times 10^{-3}$  spread in the velocity of the beam due to the source is calculated. The resultant spread due to the source  $\Delta v_s = 230$  cm/s, which is equivalent to a frequency change of  $\Delta v_s / \lambda = 230 \div (5890 \times 10^{-8}) = 3.9$  MHz. This Doppler width is well within the 10 MHz natural linewidth of the Na-D<sub>2</sub> line as is the 1 MHz linewidth of the laser beam. Therefore, the laser was capable of exciting all of the g. s.  $F = 2$  atoms in the beam and, in fact, saturating the transition. 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  $\Delta v$ , the linewidth of the atomic beam. This width is a composite of  $\Delta v_s$ , a spread due to angular divergence  $\Delta v_a$ , a spread due to power broadening  $\Delta v_p$ , and the natural linewidth of the Na-D<sub>2</sub> line  $\Delta v_n$ . At 5000 eV, the  $\Delta v$  is 55 MHz, which reduces to a spread of 37 MHz for the combined effects of  $\Delta v_s$  and  $\Delta v_a$  (after  $\Delta v_p$  and  $\Delta v_n$  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 roughly excite only  $(\Delta v_c / \Delta v) \times 31 = (30/55) 31 \approx 17\%$ , where  $\Delta v_c$  is a composite spread due to power and natural broadening. This fraction 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<sup>+</sup> 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.

Actually, not even 17% excitation can be attained because power broadening allows wing absorption to pump some of the atoms from the g. s.  $F = 2$  level to the g. s.  $F = 1$  level via the excited  $F = 2$  level. These atoms cannot then be re-excited by the laser. We will see later that only about 6% excitation could be reached.

While designing and developing the source, we decided to conduct experiments with the fused glass source and the 6% excitation that we could achieve. 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.

4. 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  $\text{Na}^*$ , 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  $\text{O}_2$ ,  $\text{NO}_2$ ,  $\text{Br}_2$ , and  $\text{I}_2$ . We settled on  $\text{I}_2$ , which has an EA = 1.72 eV. Ion pairs of  $\text{Na}^+$  and  $\text{I}_2^-$  are formed when the reactant covalent potential curve of g. s. Na and  $\text{I}_2$  crosses the product ionic curve of  $\text{Na}^+$  and  $\text{I}_2^-$ . The covalent curve of  $\text{Na}^*$  and  $\text{I}_2$  crosses the  $\text{Na}^+$  and  $\text{I}_2^-$  curve at such a large internuclear distance (and hence with negligible coupling) that no interaction and, hence, no  $\text{I}_2^-$  occurs. The  $f^*$  measured with this reaction was about 6%, as mentioned previously. We plan to publish a paper on this technique. The first experiment we conducted using a  $\text{Na}^*$  beam is briefly described next.

5. 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  $\text{Na}^*$  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  $\text{Na}^*$ -Na and Na-Na, respectively. The results were obtained by measuring  $\text{Na}^-$  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 Li- $\text{Na}^*$  collisions.

One other aspect of the  $\text{Na}^*$ -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  $\text{Na}^-$ . That is, the reaction of  $\text{Na}^*$  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  $\text{Na}^-$  product, i. e.,  $\text{Na}^-(3s^2 1S_0)$ , can

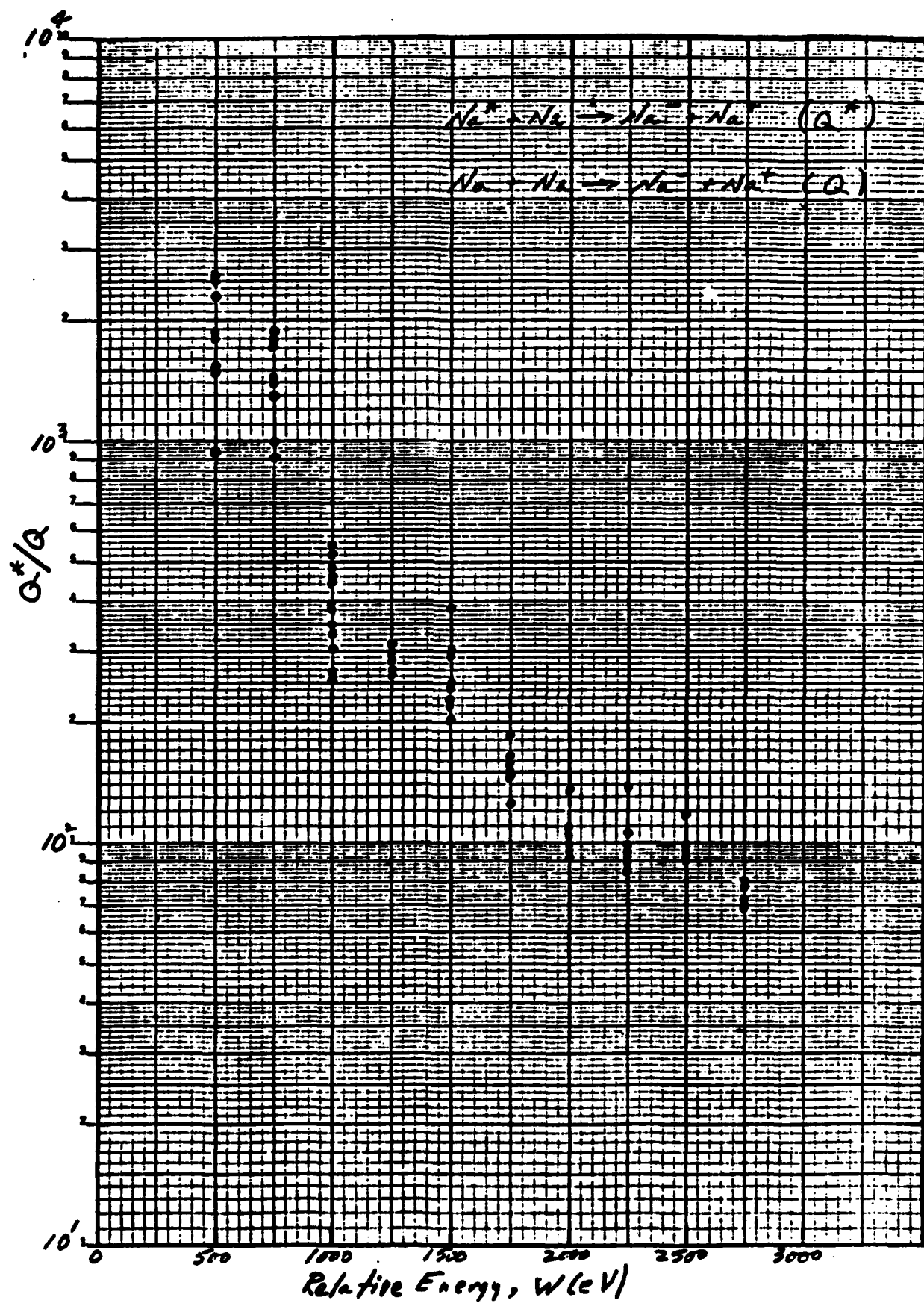


Figure 1

originate either from the reactant  $\text{Na}^*$  or  $\text{Na}$ . If it comes from  $\text{Na}$ , an electron attaches to the 3s shell, and this is a one electron process. If it comes from  $\text{Na}^*$ , not only must an electron attach itself to the 3s shell but the 3p electron of  $\text{Na}^*$  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.

6. Another preparation we made for the  $\text{Li-Na}^*$  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  $\text{Li-Na}^*$  study only a fraction of the  $\text{Na}$  beam will be excited. Most of it will be in the g. s., and the contribution to  $\text{Li}^-$  from the g. s. atoms must be known. The EA of  $\text{Li}$  and ionization potential of  $\text{Na}$  are such that the crossing radius of the covalent  $\text{Na-Li}$  and ionic  $\text{Na}^+-\text{Li}^-$  curves is 3.2 Å. This is 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  $\text{Li}^-$  produced when a fast  $\text{Li}$  beam collides with a vapor of  $\text{Na}$ .

#### Publications

1. R. H. Neynaber and S. Y. Tang, "Ion-Pair Production in Collisions of  $\text{Na}$  and  $\text{Br}$ ," accepted by *J. Phys. B*.
2. D. P. Wang, S. Y. Tang, and R. H. Neynaber, "Fractional Determination of Laser Excited Atoms in Fast  $\text{Na}$  Beams," to be submitted to *J. Phys. B*.

#### Participants

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

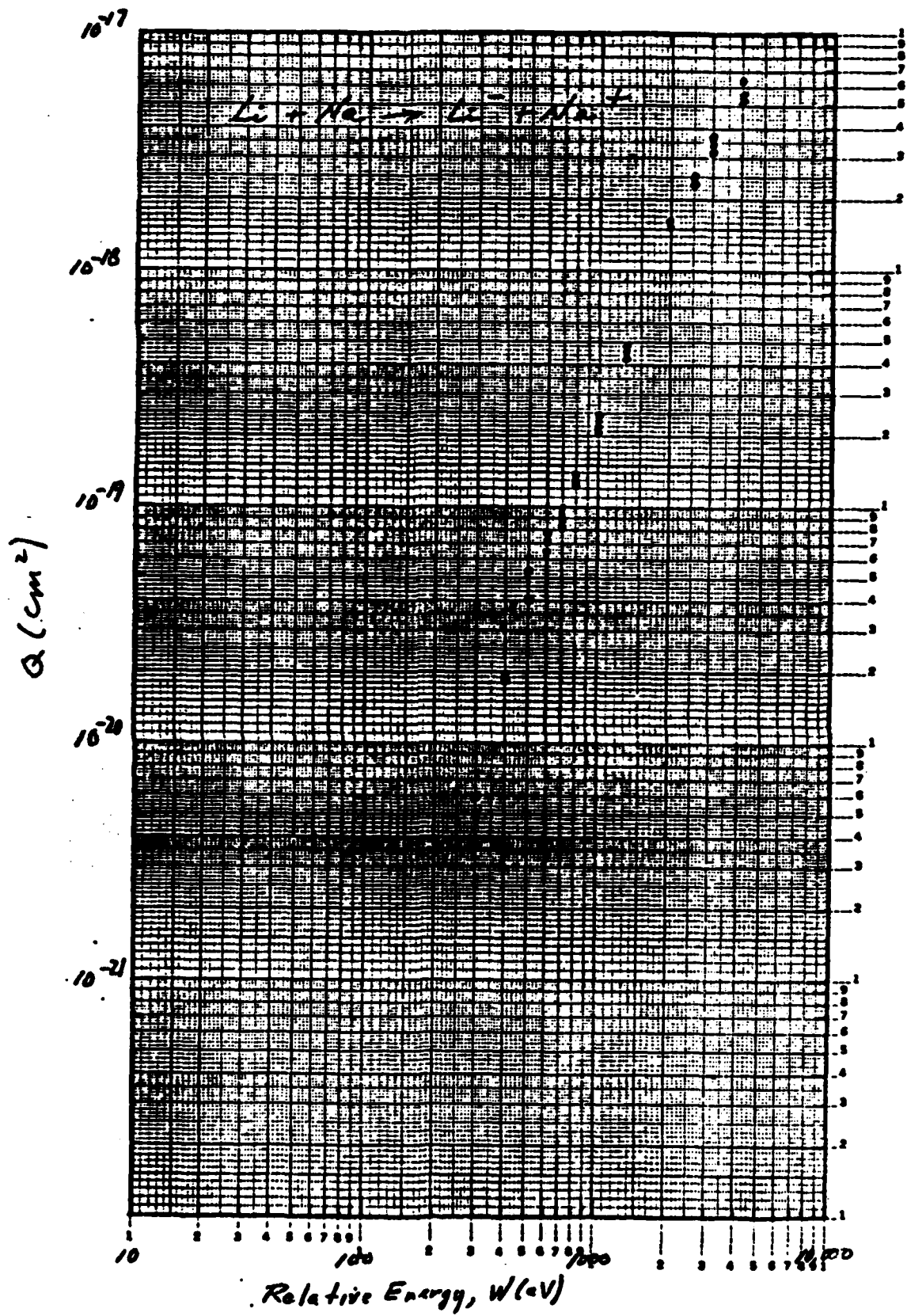


Figure 2

### Interactions

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<sup>\*</sup> 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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