the measured cross section and critical temperatures of the microbridges and the bulk critical fields are 10^5 to 10^6 times greater than the measured current-biased values. Therefore, the overall agreement between the experiment and the AL-MH theory is quite encouraging as the experiment was done at microwave frequencies while the theory was derived assuming a dc current flowing through the microbridge.

In summary the creation and collapse of localized dissipative centers in tin microbridges has been used to monitor the thermally activated motion of magnetic flux into and out of the superconducting ring. The observed switching rates are in good agreement with the theory of McCumber and Halperin. The rates were found to follow an exponential dependence in the lower-temperature region, and then with increasing temperatures the rate was found to peak and finally decrease as the temperature approached the transition temperature of the film. This is the first experimental observation of the peaking predicted by the model of McCumber and Halperin. The numerical values deduced for the free-energy barrier and the transition rate scale with microbridge cross-sectional area and are in reasonable agreement with the values prediced by Mc-Cumber and Halperin.

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¹W. W. Webb and R. J. Warburton, Phys. Rev. Lett. <u>20</u>, 461 (1968).

²J. Meyer and G. V. Minnegerode, Phys. Lett. <u>38A</u>, 529 (1972), and in *Proceedings of the Thirteenth International Conference on Low Temperature Physics*, *Boulder, Colorado*, 1972, edited by W. J. O'Sullivan, K. D. Timmerhaus, and E. F. Hammel (Plenum, New York, 1973).

³W. J. Skocpol, M. R. Beasley, and M. Tinkham, to be published.

⁴The amplitude of the microwave current bias (or the square root of the power) in the sample under these conditions varied with temperature as $(T_c - T)^{3/2}$. This temperature dependence is the same as the observed mean-field critical-current variation in our samples and thus the ratio of bias current to effective critical current was constant. Therefore $\Omega'(I/I_c)$ and $F'(I/I_c)$ as given in Eq. (1) were measured at a constant value of the ratio I/I_c . A further discussion of this point will appear in a later publication.

⁵J. S. Langer and V. Ambegaokar, Phys. Rev. <u>164</u>, 498 (1967).

 $^6 \mathrm{D.~E.~McCumber}$ and B. I. Halperin, Phys. Rev. B <u>1</u>, 1057 (1970).

⁷The equation for the rate prefactor Ω' in the presence of a current is given by Eqs. (2.8), (3.38), and (4.36) of Ref. 6 and the free-energy parameter F' is derived from Eqs. (3.25) and (3.13) of Ref. 5.

⁸The "first step" refers to the increment in the sample dissipation encountered at lowest microwave input power; the "second," "third," "fourth," etc. steps refer to subsequent steps encountered in this sequence as the microwave power is increased. A detailed discussion of the properties of these steps will be discussed in another publication.

Temperature-Modulated Reflectance of Gold from 6 to 35 eV

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The thermoreflectance spectrum of gold has been measured in the 6-35-eV region at about 200 K by using synchrotron radiation. Considerable sharp structure exists throughout this region, showing that lifetime broadening is not large enough to preclude the extension of high-resolution modulation techniques to at least 35 eV. The structure arises from interband transitions of the 5d electrons and from plasmons.

The contributions of modulation spectroscopy to the understanding of the band structure of solids have proven to be fundamental. This technique is the only one capable of displaying contributions to the optical spectra arising only from localized regions in the Brillouin zone, such as

critical points in the band structure.^{1,2} Until now, modulation spectroscopy, with few exceptions,^{3,4} has been limited to the spectral region from the infrared to about 10 eV, for lack of suitable radiation sources. Recently, intense synchrotron radiation from the extremely stable beam in the storage ring Tantalus 1, operated by the Physical Sciences Laboratory of the University of Wisconsin,⁵ has made high-sensitivity modulation measurements possible. The thermomodulation spectrum of gold reported in this Letter is the first spectrum above 10 eV resulting from the application of an external modulating field to a sample. The spectrum shows a great deal of structure, some of which is remarkably sharp. It has been necessary to use a spectral bandpass of less than 100 meV at 25 eV to reveal the details. This spectrum demonstrates that the high resolution possible with modulation spectroscopy will be valuable for probing the band structure of materials for away from the Fermi level.

The samples were 400-Å gold films evaporated on quartz or sapphire substrates. They were self-heated with a unipolar 1.6-Hz square-wave current supplying 3.75 W peak power. Other frequencies and heating powers were used with the spectra scaling predictably. The optical system was basically that reported earlier,⁶ the principal change being a smaller, simpler sample chamber. The chamber had a liquid-nitrogen cold finger and was ion pumped to a base pressure of less than 10⁻⁸ Torr to avoid condensation on the sample. The avarage sample temperature was about 200 K and the modulation was 0.2-0.5 K. Radiation was incident on the sample at 30° with primarily p polarization. In the 6 to 25 eV range a conventional sodium salycilate photomultiplier combination was used. From 12 to 35 eV an EMI model 9603/2B electron multiplier provided a great increase in sensitivity along with the rejection of long-wavelength stray light. The dc signal (proportional to the reflectivity R) was monitored with an electrometer while the ac signal (proportional to the change in reflectivity ΔR) was detected by a lock-in amplifier. To provide long integration times the lock-in was used with a short time constant and the output was fed to a voltage-to-frequency converter and counted for 60 to 120 sec. The noise limit in $\Delta R/R$ was photon shot noise and varied from 5×10^{-6} at the peak of the monochromator output to 5×10^{-5} at the highest energies. The sample-to-sample variations in the thermoreflectance spectrum were very small and appeared as magnitude effects with no change in structure.

Figure 1 shows the thermoreflectance spectrum of gold as a function of energy E from 6 to 35 eV, as well as our measured reflectivity, dR/dE, and d^2R/dE^2 . The most striking feature of Fig. 1 is the very rich structure in the thermoreflectance



FIG. 1. (a) Thermoreflectance of Au at 200 K, 30°, p polarization. (b) Reflectivity of Au at 30°, p polarization. (c) First derivative dR/dE (dashed line) and second derivative d^2R/dE^2 (full line) calculated from the reflectivity shown in (b).

spectrum up to 35 eV. This result contrasts with a widespread feeling that broadening at higher energies due to shorter lifetimes of the excited states would tend to wash out any structure and make modulation spectroscopy unimportant at higher energies.

Our thermoreflectance spectrum reproduces the data of Scouler⁷ very well from 6 to 10 eV. The interpretation of this portion of the spectrum has been carried out by Christensen and Seraphin,⁸ who found good agreement between the energies at which the modulated reflectance of gold shows structure and the energies they have calculated for critical-point transitions. There has been no detailed interpretation, however, of the optical properties of gold above 10 eV although the optical constants are well known.⁹⁻¹¹

Structure in the 10-35-eV thermoreflectance cannot be due either to transitions involving core states, since the highest core level in gold lies about 54 eV below the Fermi level,¹² or to transitions terminating at the Fermi level, which VOLUME 33, NUMBER 11

lies 9.5 eV above the bottom of the conduction band.⁸ Thus all the structure in $\Delta R/R$ will arise from interband transitions originating in the 5dbands or at the Fermi level, or from plasmons. There are two plasmonlike peaks in $\text{Im}(-1/\tilde{\epsilon})$ for Au at 25.8 and 32.6 eV.^{11,13} Each of these should yield a characteristic structure in $\Delta R/R$.¹⁴ The beginning of the structure below 25 eV is obscured by structure due to interband transitions, but the positive peak at 28.5 eV and the negative peaks at 32 eV are of the expected shape and at the expected positions. Band shifts upon thermal expansion cause modulation spectra resembling dR/dE. Interband transitions are broadened by phonon scattering, the temperature dependence of which gives rise to a thermoreflectivity spectrum resembling d^2R/dE^2 . Thermal broadening of the Fermi function can also give spectra resembling d^2R/dE^2 , but transitions originating on the Fermi surface should contribute little to the thermoreflectivity above 10 eV, since no regions of high joint density of states are cut by the Fermi level. Given the general agreement of $\Delta R/R$ and d^2R/R dE^2 , between 10 and 25 eV, thermal broadening is the principal cause of the thermomodulation. Then the energies of the negative peaks in $\Delta R/R$ should correspond roughly to critical-point thresholds in the joint density of states¹⁵ for transitions originating in the 5d bands and the lower part of the 6s band.

In Table I we have correlated structures in the thermoreflectance with critical points at Γ , K, and W and along Q which can be identified in the bands calculated by Connolly and Johnson.¹⁶ Several critical points are almost degenerate in energy and overlap to give broader structures. At this stage, it is not possible to discriminate between them. Note, however, that the structures in the 10–18 eV region are relatively weak and broad. Some of the transitions appear as multiplets, having the same final state but originating in different d bands. A small shift of the energy of the final state brings the theoretical values of the transition energies into closer agreement with the experimental values for the entire multiplet, as shown in the fourth column of Table I. Above 18 eV there are five strong, sharp structures. Structure is expected from the M_0 criticalpoint transitions $\Gamma_{8^+} \to \Gamma_{7^-}, \ \Gamma_{7^+} \to \Gamma_{7^-},$ and Γ_{8^+} $\rightarrow \Gamma_{7-}$, but only two peaks are at the calculated positions. If the energy of the Γ_{7} final state is increased 1.3 eV the predicted transitions occur at 20.1, 21.2, and 22.5 eV, in close agreement with the sharp structures at 19.9, 21.2, and 22.6

TABLE I. Structure in the thermoreflectance spectrum of Au and its assignment to critical-point transitions identified from the calculated bands of Ref. 16 at some symmetry points and directions in the Brillouin zone. The suffix labeling the states corresponds to the number of the band counted from the lowest band. By applying shifts of 0.2, 0.3, and 1.3 eV to final states Q_7 , W_9 , and Γ_{7-} , respectively, the calculated positions in the third column have been obtained.

| | TUEO | ENERGIES (eV) SHIFTED | |
|---------------------------------------|---------|-----------------------------|----------|
| ASSIGNMENT | RETICAL | RETICAL | MENTAL |
| $Q_2 \rightarrow Q_7$ | 8.7 | 8.9 | 9.0 |
| K ₄ → K ₇ | 8.9 | J | J |
| $K_3 \rightarrow K_7$ | 9.7 | | 10.0 |
| $K_4 \rightarrow K_8$ | 10.4 | 1 | 10.6 |
| $Q_1 \rightarrow Q_7$ | 10.4 | 10.6 | 10.0 |
| $W_5 \rightarrow W_9$ | 11.4 | 11.7 | 11.7 |
| $W_2 \rightarrow W_7$ | 11.7 | J | , |
| $W_4 \rightarrow W_9$ | 12.8 | 13.1 | |
| $W_2 \rightarrow W_8$ | 13.2 | } | 13.2 |
| $K_2 \rightarrow K_8$ | 13.3 |) | |
| $W_3 \rightarrow W_9$ | 14.4 | 14.7 | 14.7 |
| $\Gamma_{8+} \rightarrow \Gamma_{7-}$ | 18.8 | 20.1 | 19.9 |
| $\Gamma_{7+} \rightarrow \Gamma_{7-}$ | 19.9 | 21.2 | 21.2 |
| $\Gamma_{8+} \rightarrow \Gamma_{7-}$ | 21.2 | 22.5 | 22.7 |
| - | - | - | 17.3 |
| - | - | - | 21.8 |
| - | - | - | 24.4 |

eV.17

The fact that there is a pronounced peak in the reflectivity between 18 and 27 eV indicates that considerable oscillator strength is concentrated in this region. (See the ϵ_2 sum rule plot in Fig. 4 of Ref. 11.) The 5*f* levels in Au contribute to the conduction bands at about 20 eV above $E_{\rm F}$.¹⁶ Actually, the states near Γ_{7-} are mostly of *f* type. Transitions from *d* to *f* states generally have large oscillator strengths, so that the M_0 critical-point transitions we assigned above are expected to be strong, and may account for the large strength of the 18–23 eV peak in ϵ_2 .¹¹ The presence of the "5*f* bands" may explain some of the anomalies observed in ultraviolet photoemission.¹⁸

We have shown that modulation spectroscopy can be extended with the necessary high resolution up to at least 35 eV. Available band-structure calculations, based on Fermi-surface and ultraviolet-photoemission-spectroscopy measurements, give good results for the filled parts of the conduction bands and account for the lowerenergy interband absorption, but they do not explain satisfactorily all of our higher-energy data. The thermoreflectance spectrum presented in this Letter provides a great amount of new information, mostly concerning final states far above the Fermi level, information which is necessary for future improved calculations of highly excited states. We also suggest that evaluation of some dipole matrix elements would enable a better understanding of the optical and ultraviolet photoelectron spectra.

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³R. H. Willens, H. Schreiber, E. Buehler, and D. Brasen, Phys. Rev. Lett. <u>23</u>, 413 (1969); R. H. Willens and D. Brasen, Phys. Rev. B <u>5</u>, 1891 (1972).

⁴W. Zierau, in Proceedings of the International Sym-

posium for Synchrotron Radiation Users, Daresbury, England, 1973, edited by G. V. Marr and I. H. Munro (Daresbury Nuclear Physics Laboratory, Daresbury, England, 1973).

⁵E. M. Rowe and F. E. Mills, Particle Accel. $\underline{4}$, 211 (1973).

⁶C. G. Olson and D. W. Lynch, Phys. Rev. B <u>9</u>, 3159 (1974).

⁷W. J. Scouler, Phys. Rev. Lett. <u>18</u>, 445 (1967).

⁸N. E. Christensen and B. O. Seraphin, Phys. Rev. B $\underline{4}$, 3321 (1971).

 ${}^{\widehat{9}}W$. C. Walker, O. P. Rustgi, and A. L. Weissler, J. Opt. Soc. Amer. 69, 471 (1959).

¹⁰L. R. Canfield, G. Hass, and W. R. Hunter, J. Phys. (Paris) 25, 124 (1964).

¹¹B. R. Cooper, H. Ehrenreich, and H. R. Philipp, Phys. Rev. 138, A494 (1965).

¹²J. A. Bearden and A. F. Burr, Rev. Mod. Phys. <u>39</u>, 125 (1967).

¹³J. L. Robins, Proc. Phys. Soc., London <u>78</u>, 1177

(1961); M. Creuzburg, Z. Phys. <u>196</u>, 433 (1966). ¹⁴Ref. 1, pp. 120-121.

¹⁵R. Rosei and D. W. Lynch, Phys. Rev. B <u>5</u>, 3883 (1972).

¹⁶J. W. D. Connolly and K. H. Johnson, Massachusetts Institute of Technology Solid State and Molecular Theory Group Report No. 72, 1970 (unpublished), p. 19.

¹⁷The splitting of the levels at Γ of the 5*d* bands as calculated by N. V. Smith, Phys. Rev. B <u>9</u>, 1365 (1974), agrees with the experimental splittings within the instrumental bandpass (0.1 eV). We have chosen, however, to use the bands of Connolly and Johnson (Ref. 16) because they extend to higher energies. [The calculation of Christensen and Seraphin (Ref. 8) does not include as many bands through Γ near 1.8-2.0 Ry.]

¹⁸J. Freeouf, M. Erbudak, and D. E. Eastman, Solid State Commun. <u>13</u>, 771 (1973).

Stability of Electron-Hole Drops in Germanium under Uniaxial Stress

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The cohesive energy of the electron-hole drop in germanium at 1.5 K is determined as a function of uniaxial stress, both along $\langle 100 \rangle$ and along $\langle 111 \rangle$ directions, by the method of cyclotron-resonance monitoring at 35 GHz. On application of stress, it starts decreasing from the value of 14.5 K at zero stress, but has an air of persisting to have a finite value at a considerably high-stress region.

At low temperatures, condensation of excitons into electron-hole drops occurs in germanium.¹ The ground-state energy, or cohesive energy, of such a condensed system has recently been studied by Combescot and Nozières,² Brinkman and Rice,³ and Vashishta, Bhattacharyya, and Singwi.⁴ Their results all show that the complexity of the actual band structure in germanium is favorable for a stable drop formation. It would be of interest, accordingly, to modify the band structure by applying a uniaxial compressive stress which lifts the degeneracies of the conduction and va-

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¹M. Cardona, *Modulation Spectroscopy* (Academic, New York, 1969).

²Surface Sci. <u>37</u> (1973).