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Publication Date

1975-09-01

LBL-4563

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STUDY OF ATOMIC SCATTERING FACTORS FROM THE CRITICAL VOLTAGE OF ORDERED ALLOYS - APPLICATION TO Cu_3Au

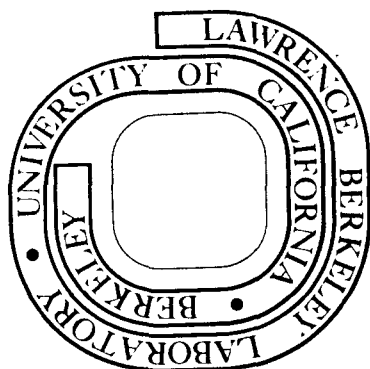A. Rocher, R. Sinclair, and G. Thomas

September 1975

LBL-4563

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DISCLAIMER

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The critical voltage (V_c) effect, first discovered by Nagata and Fukuhara (1967) and Uyeda (1968), has already been used to determine experimental values of the Atomic Scattering Factor (ASF). The main interest of this study is to yield complementary information about ASF and Debye temperatures from data on an ordered alloy, Cu3Au, using the technique developed by Rocher and Jouffrey (1972, 1974).

For ordered alloys reflections with low values of $\sin \theta / \lambda$ are allowed, e.g. 100 and 110 are reflections of the Ll$_2$ structure of Cu3Au whereas the first reflection of the disordered f.c.c. lattice is 111. The superlattice interaction potentials V_{100} and V_{110} also perturb dynamical effects such as critical voltage and it is then possible to determine their values as a function of V_c.

Methods of caluclations

Two eigenvalues of the fundamental equation are equal at the critical voltage, which is the criterion taken for calculations of V_c. The difference between the two eigenvalues is to a first approximation a linear function of the accelerating voltage. The precision of the calculation has been limited to one kilovolt. Computation times are reasonable with about five iterations, for the multi-beam value, from the value determined from three beam theory. By comparison, experimental results are only accurate to within a few kilovolts.

Choice of parameters

Critical voltages are calculated with a limited number of parameters. Compared with the experimental values (least squares method), they permit the determination of the real values of the ASF of an atom in pure metals and the Debye temperature of an alloy.

a) Atomic Scattering Factor

L. E. Thomas et al. (1974) showed that the Doyle and Turner (1968) ASF in gold yields calculated critical voltages very close to the experimental results for angles larger or equal to that for the 200 reflection.

The experimental values of the copper ASF are given by Rocher and Jouffrey (1972, 1974) for reflections with angles larger or equal to that for the 111 reflection.

The ASF of alloys are calculated with the assumption that the A atom in the pure A metal and the A atom in the $A_n B_{1-n}$ alloy have the same ASF. Then for Cu3Au ASF Fg are correctly determined from ASF values known for copper and gold. In the formulation used by Doyle and Turner (1968):

$$Fg = \Sigma_{i=1-4} A_i \exp - B_i g^2 \qquad (1)$$

The last three terms, giving the variation for large angles, can be considered as correct. For copper and gold the first term only need be corrected in order to yield good agreement between experimental values and calculated values of V_c for the fully ordered alloy.

b) Debye Temperature

Shirley et al. (1975) give an equation to define the Debye temperature of an alloy, θ, a function of the mole factions M_A and M_B of A and B atoms, μ_A and μ_B their atomic weight and θ_A and θ_B their pure metal Debye temperatures. The relation is written:

$$(M_A\mu_A + M_B\mu_B)\theta^2 = M_A\mu_A\theta_A^2 + M_B\mu_B\theta_B^2 + (\tau-1)\,(1-\alpha)\,M_AM_B(\mu_A\theta_A^2 + \mu_B\theta_B^2) \qquad (2)$$

where α is the nearest neighbour Cowley Warren short range order parameter (Cowley 1950) and τ is a characteristic constant of the interatomic forces, which is established from the critical voltage.

Results

The experimental results used in this paper are those of Thomas et al. (1974) and Sinclair et al. (1975). Accurate measurements of the 440 critical voltage can not be obtained because of the high irradiation damage rates produced by the high energy electrons at ~1400kV and only an approximate comparison is possible for this reflection.

The ordered alloy permits the determination of the coefficient A_1 for copper and gold, since $\alpha = 1$ and the Debye temperature is well known from equation (2) and macroscopic data.

The values of A_1(Cu) and A_1(Au) shown in Table I were calculated using 400 critical voltages taken at two different temperatures, with the 222 critical voltage used as a check.

Table I

	A_1	V_{100}	V_{200}
Cu	$2.75 \pm 0.1\text{Å}$	$18.5 \pm 0.2V$	$11.00 \pm 0.05V$
Au	$2.15 \pm 0.2\text{Å}$	$24.4 \pm 0.4V$	$17.25 \pm 0.05V$

These data show that the ASF of an atom within the gold lattice and that of a single gold atom have almost the same values. However, for copper the scattering by a free atom and the scattering by an atom within the crystal are quite different. It is not known at this stage whether this arises from unreliable assumptions in the Hartree-Fock method or from the influence of the lattice on the scattering by an individual atom.

Table II shows the experimental and theoretical values of critical voltage calculated from these ASF's and the Debye temperatures established from equation 2. It can be seen that we have exact correspondence between the two, within the limits of experimental error. (The uncertainties in the calculated V_c result from experimental uncertainties of the important parameters). Thus our approach is an improvement over the previous calculations of Thomas et al (1974) for Cu_3Au by which the calculated values exceeded the experimental values by about 10%. The basic reason for the current improvement is due to the reliable values of ASF.

Table II

Fully Ordered Cu_3Au		L.E. Thomas et al.	Sinclair et al.	Calculation
$\alpha = 1$	222	$175 \pm 3KV$		$175 \pm 2KV$
$\theta = 261K$	400	$425 \pm 3''$	$420 \pm 10KV$	$422 \pm 3''$
$\tau = 293K$	400 (T=461K)	$364 \pm 3''$		$363 \pm 3''$
	440		> 1400 ''	$1470 \pm 10''$
Disordered Cu_3Au				
$\alpha = 0$	222	$166 \pm 3''$		166 ± 2 ''
$\theta = 193K$	400	$381 \pm 3''$	$360 \pm 10''$	381 ± 3 ''
$\tau = 293K$	440		≈ 1300 ''	$1285 \pm 10''$

The variation of 222 critical voltage with degree of order cannot be explained by a variation of ASF (which is the same for both ordered and disordered states) but is due to the variation of Debye temperature on ordering as described by Shirley et al (1975) using equation 2. The coefficient τ was thus found to be 0.70 ± 0.04 for disordered Cu_3Au ($\alpha = 0)_x$ from the 222 and 400 critical voltages. This may be compared with the value $^x\tau = 0.72$ found for Fe-Cr alloys by Shirley et al (1975).

Acknowledgements

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